ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

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Isotopes and Radiation Technology

Vol. 7, No. 4 Summer 1970

Contents

I Isotope Production and Development

363 **Ad Hoc Panel Report on the National Uses and Needs for Separated Stable Isotopes**
364 Scientific and Technical Uses and Needs
374 Administration of Programs
374 Conclusions
376 Recommendations
378 **The Chemistry of the Electromagnetically Separated Stable Isotopes**

II Isotope Technology Development

381 **Industrial Measurement Considerations for Use of Low-Energy Photons from Nuclear Sources** *Howard J. Evans*
382 Sources of Low-Energy Photons
384 Principles—Interaction of Photons with Materials
386 Detectors
391 Applications
392 Future
396 **Development of Some Radioisotope Procedures for Measuring Water Movement in Trees** *P. W. Owston, J. L. Smith, and H. G. Halverson*
397 Literature Review
398 Feasibility Test
400 Field Test of Technique
402 **Old Bremsstrahlung University: Two Decades of Training in the Nuclear Sciences** *John F. Yegge and Grover Smithwick*
402 Special Training Courses
403 Mobile Radioisotope Laboratories
404 International Exhibits
405 Recent Achievements
406 Plans for the Future
407 **Radioactive Tracer Technique for Laboratory Corrosion Studies** *Harry J. Close*
407 Experimental
408 Results
410 Conclusions
411 **Use of ^{252}Cf for Mineral Exploration: Comparison with Accelerators for In Situ Neutron Activation of Silver** *F. E. Senftle, P. W. Philbin, and P. Sarigianis*
411 Source Descriptions
414 Results
418 Comparison of Sources
419 **Applications and Potential Applications of Radioisotopes to Melt-Shop Steelmaking Operations** *W. B. Tucker*
419 The Melt Shop
419 Processes and Materials
424 Isotope Applications to Melt-Shop Raw Materials

XUM

419 **Applications and Potential Applications of Radioisotopes to Melt-Shop Steelmaking Operations** *(Continued)*
426 Isotope Applications to Melting and Refining
429 Isotope Applications to Casting
431 Miscellaneous Melt-Shop Applications
432 Conclusion
434 **Inspection of Hand-Grenade Fuses with a Radioactive Isotope**
435 Radioactivity Required for a Given Inspection Rate
435 Test Results
436 Safety Considerations
436 **Use of Radioisotopes in Detector Calibrations** *J. H. McCrary*
437 Source Requirements
437 Possible Sources
439 Experimental Procedure
440 **Gamma-Ray Weld-Inspection System** *D. M. Payne*
440 Weld-Inspection System

III **Isotopic Heat and Power Development**
443 **SNAP-27 on the Moon** *D. L. Prosser*
443 Power Source
446 Capsule
448 **Radioisotopic Heat Source on the Moon** *D. P. Kelly and V. L. Avona*
449 **Isotopic Energy Systems for Underseas Application**

IV **Isotopes Around the World**
452 **Radioisotope Program in India** *V. K. Iya and R. G. Deshpande*
452 Radioisotope Production
456 Radioisotope Applications
457 Conclusion
458 **Portable ^{170}Tm Gamma-Radiography Unit for Light Metals**
459 **Sealed Radioisotopic Sources at Nuclex 69**
461 **Gamma Irradiator Using Spent Fuel Elements**
462 **Economics of Industrial Radiation-Chemical Sulfonation of Hydrocarbons**
462 **10 Years of Radioisotopes in East Germany**
463 **Quick Balance of Mercury in Electrolytic Cells by Using Radioactive Mercury**
463 *Nuclear Methods in the Aviation Industry*
464 **Determination of Heavy-Oil Heat Value with Neutrons**
464 **Coal-Ash Measurement with Two Radioisotopic Radiation Sources**
465 **Survey of Radioisotope Applications in Latin America**
G. B. Baro and J. G. Flegenheimer
465 Uses
468 Future

V **Miscellaneous**
469 **Availability of Isotopes and Services**
471 **AEC Activities**
473 **General**

Index
479 **Volume 7**

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

Insofar as feasible all references are checked by the editors, using the library facilities of Oak Ridge National Laboratory, Oak Ridge Associated Universities, and the University of Tennessee. Occasionally, however, the cited material is not available; such cases are identified by an asterisk (*).

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, and F. J. Miller, *Editors*
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OAK RIDGE NATIONAL LABORATORY

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TECHNICAL PROGRESS REVIEWS

The United States Atomic Energy Commission publishes the Technical Progress Reviews to meet the needs of industry and government for concise summaries of current nuclear developments. Each journal digests and evaluates the latest findings in a specific area of nuclear technology and science. *Nuclear Safety* is a bimonthly journal; the other two are quarterly journals.

Isotopes and Radiation Technology
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Nuclear Safety
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Reactor Technology
Prepared in cooperation with the American Nuclear Society

All are available from the U. S. Government Printing Office. See the back cover for ordering instructions.

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Other U. S. Government agency reports identified in this journal are generally available from CFSTI.

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Private-organization reports should be requested from the originator.

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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Ad Hoc Panel Report on the National Uses and Needs for Separated Stable Isotopes

Editor's Note: The following report presents the findings and recommendations of the ad hoc panel on the National Uses and Needs for Separated Stable Isotopes. This edited version of the National Research Council report was prepared by the Isotopes Information Center. Neither the members of the panel nor the National Research Council has edited or approved the present document; the National Academy of Sciences–National Research Council publication, "A Report on National Uses and Needs for Separated Stable Isotopes," dated July 29, 1968, should be referred to as the original source. The panel consisted of:

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Rice University
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Abstract: *This ad hoc panel report surveys the role that the U. S. Atomic Energy Commission's stable-isotope separations program has had in advancing nuclear sciences and reviews the program in terms of present and future needs. Past, present, and future uses of separated stable isotopes in basic and applied nuclear physics, nonnuclear physics, chemistry, earth sciences, and biology and medicine are discussed. The panel recommends the acceptance and continuance of the program as a national resource of great value to the United States.*

The large-scale separation of isotopes* started during World War II because of a need for fissionable material. In January 1947 the U. S. Atomic Energy Commission (AEC) assumed control of a program started about a year earlier under the Manhattan Engineer District for the separation of stable isotopes for basic and applied nuclear science studies. The program has been very successful and has resulted in significant advances in the nuclear sciences. This report surveys the role that the isotope separations program has had in these advances and reviews the AEC program in terms of present and projected needs of the scientific community.

The AEC separations program has been centered mainly at the calutron (electromagnetic separation)

*Throughout this report the term "separated isotopes" refers to separated (enriched) stable isotopes.

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facility* at Oak Ridge, Tenn., but contributions (gaseous separations) have been made by Mound Laboratory, Miamisburg, Ohio, as well as by some other AEC laboratories and contractors. Separated isotopes are distributed mainly by sale from Oak Ridge and from Mound Laboratory; however, the AEC has established an Isotopes Pool for loan of isotopes for approved nondestructive uses

The present report is concerned principally with stable-isotope separation and use; however, the distinction between stable and radioactive isotopes is arbitrary when the radioactive half-life is very long, as is the case with some naturally occurring radionuclides, or when an enriched radioisotope is prepared by using a separated stable isotope as a target material for a nuclear transmutation.

The most obvious needs for separated isotopes are for use in studies of the nuclear properties of atoms. This report first surveys uses in nuclear physics studies, including both basic and applied aspects of nuclear physics and engineering and then reviews the uses in nonnuclear physics, chemistry, earth sciences, and biology and medicine.

Since the association of nuclear science with other sciences, either directly or indirectly, is largely through the use of separated stable isotopes, their uses are described to show their vital and essential nature and to emphasize the unequivocal need for their continued availability. These uses involve not only small quantities for tracer applications but also larger quantities for Mössbauer and magnetic-structure studies. In many instances radioisotopes are used in medical studies of their radiological effects, and these isotopes frequently are prepared with high specific activity by activation of separated stable isotopes. In some cases no suitable radioisotope exists for tracer studies of a particular element; thus one must resort to isotope-dilution techniques, using enriched stable isotopes and conventional methods of isotopic analysis (mass spectrometry, activation analysis, etc.).

In the report both the past and current uses as well as the future needs for separated stable isotopes in the various scientific disciplines are discussed. The administration of the AEC programs at Oak Ridge and at Mound Laboratory is briefly reviewed, and some general conclusions and recommendations of the panel are given.

*The calutron facility is located in the Y-12 Plant area but is operated by the Isotopes Division of the Oak Ridge National Laboratory (ORNL). Both ORNL and the Y-12 Plant are operated by Union Carbide Corporation, Nuclear Division.

Scientific and Technical Uses of and Needs for Separated Stable Isotopes

Isotopes Pool

Because of the need for large samples of separated isotopes, primarily for measurements of neutron cross sections but to some extent for other nuclear physics measurements, the Nuclear Cross Section Advisory Committee suggested in 1956 the creation of what is now commonly called the "Isotopes Pool." Their report made specific recommendations as to (1) the minimum size of such a loan pool, based on the sample size needed for neutron-cross-section measurement techniques at that time, and (2) the means of filling the needs for the larger quantities of isotopes. Samples of separated isotopes, sometimes in very special chemical and physical states, were to be loaned by the AEC for nondestructive use. The committee also recommended that additional calutrons for stable-isotope separation be made available and that a total of 183.9 gram atoms of nearly pure nuclides be separated for the pool by about 1964. Clearly it is now time to review this program and to attempt to establish up-to-date objectives.

The Jan. 31, 1968, inventory report of stable isotopes showed 454 gram atoms—about 140 in the active inventory, which is offered for sale, and 314 in the pool, which contains rather large samples that must be returned, after use, without any important loss and without introduction of appreciable radioactivity into the sample. The total is well ahead of the minimal objectives established in 1957, but some of the elements discussed in the 1957 report have been separated in larger amounts than recommended and others in less. The excess of the currently available pool material over the minimal 184 gram atoms recommended in 1957 is represented by an oversupply of nuclides with a very high natural isotopic abundance, while there is an undersupply of most nuclides of relatively low natural abundance. The pool is therefore still appreciably short of its goal of 13 years ago.

It would be possible in a few years to fulfill the 1957 objectives if top priority were given simply to making up this inventory. However, if the program continues to give first priority to satisfying requirements for separated isotopes needed for studies of the newer ideas in the scientific community, the arbitrary goals of 1957 may never be reached. Thus the pool is in no danger of saturation from this standpoint. Other considerations show that these earlier goals were minimal.

The 1957 objectives were set for measurements of total neutron cross sections by using crystal spectrometers and fast-neutron choppers and for measurement on the scattering of homogeneous-in-energy neutrons produced by Van de Graaff generators with either continuous or pulsed beams. Most of the measurements, using methods known in 1957 and made with practical quantities of available separated isotopes, have now been completed. Since 1957, powerful new methods for measuring neutron absorption, transmission, and scattering have been discovered. The principal new development is the production of very strong beams of neutrons, either by underground nuclear bomb explosions or by new types of accelerators with intense beams of electrons or deuterons. Since these beams may burn out the target, limited target areas and the resulting heat transfer problems must be alleviated by increasing the area bombarded by the beam. Consequently any sample that can intercept the neutrons from such a beam must be much greater in area than those needed now. Samples likely to be needed to make full utilization of these new neutron spectrometers will probably increase by a factor of more than 2 over those contemplated in 1957. Furthermore, most of the important measurements that can be performed satisfactorily with naturally occurring isotopic mixtures have already been completed. In the near future a greatly increased demand for separated isotopes to be used in making nuclear measurements can be expected. Whether or not the new series of measurements will be as extensive as those of the older program is not clear, but there is reason to believe that it will, and this, in turn, emphasizes the need for continuing the stable-isotope separations program at the present rate for sometime at least. Thus we see that the earlier goals were indeed minimal.

Isotope Separations Technology

Second-Pass Separations. The purity of some separated isotopes at present is not completely satisfactory for some measurements; therefore a "second pass" through the calutron for certain isotopes is indicated. In principle, this operation does not imply very large losses, but a considerably more expensive process of cleaning the calutrons is involved. Largely because the output of the calutrons is in such high demand for an ever-increasing number of nuclides, the development of the second-pass technique and the accumulation of an inventory of adequately purified isotopes of barium, strontium, titanium, and other elements have barely been started. Even if all need for increased quantities of isotopes in the pool should vanish, continued full-time use of the present calcutrons would be required for many years for such second-pass work for the accumulation of a pool of more highly purified separated isotopes.

The needs for samples for inelastic-scattering measurements have perhaps been analyzed more carefully than any of the others; the conclusion is very clear—⅔- rather than ⅓-gram-atom quantities will be required for the new techniques. Furthermore, the insufficiently large samples are primarily of elements whose isotopes cannot yet be separated very efficiently. Since the program has been running for about 8 years and has not yet satisfied the 1957 pool estimate based on ⅓-gram-atom samples, there seems to be no reason to decrease the separation effort at this time.

Research and Development Effort. The calutron program is not devoted entirely to routine production by established methods. A very considerable research and development program is conducted to improve the electromagnetic separation process. Although the demand for the calutron product shows no sign of slackening, the techniques may well have matured sufficiently to suggest that the research and development program might be curtailed. However, the lack of uniformity in the success of separation of all elements up to this time has, in fact, increased the demand for the isotopes of the less easily separated elements. For instance, a recent projection of future needs for neutron-cross-section measurements that are of interest to space science and cosmology indicates a pressing need for isotopes of Hg, Pt, Os, and Pd, none of which is easily separated by existing methods. Development will need to continue, particularly on ion sources.

Basic Nuclear Physics

Past and Present Uses. Since the details of nuclear spectrums, decay, transmutations, and reactions are specific to isotopic composition, the study of these basic nuclear characteristics is facilitated by using separated stable isotopes. Study of the sales pattern shows that in FY 1964 about 45% of the stable isotopes were used for basic nuclear studies. In FY 1967, sales had more than doubled for this use, although the fractional sales dropped to 40% because of increased usage in other fields.

Where the natural abundance of an isotope is small, the measurement of its nuclear properties may be impossible unless the separated (enriched) isotope is used. Moreover, the study of nuclear properties of a

XUM

particular isotope in the natural state, even where its isotopic abundance is large, may be impossible with a naturally occurring specimen or inaccurate when interferences from less abundant isotopes of that element occur. Thus for a large number of basic nuclear measurements, isotopically enriched samples are required. Futhermore, since nuclear structure, spectroscopy, and reaction studies are concerned with the systematics of all the isotopes of an element, separated samples of nearly all isotopes have been and continue to be required. A survey of publications in the *Journal of Nuclear Physics* for the years 1966 to 1968 shows that of 1071 papers published in the general area of basic experimental nuclear physics, 477 (or 44.5%) used isotopically enriched materials. These studies covered virtually all the available separated stable isotopes.

Much knowledge of the nature of nuclear forces and of the selection rules governing nuclear reactions has been gained in recent years from studies using separated isotopes. An interesting recent example has been the recognition of "isobaric analog states" in nuclei. This new information has advanced considerably the understanding of nuclear forces and nuclear structure and was made possible by experiments such as proton scattering from targets of separated stable isotopes.

Various models of nuclei have been developed to provide a systematic description of some of the experimentally determined nuclear properties. The shell structure of nuclei, described at least partially by various shell models, is based on nuclear parameters such as the mass and angular momentum of specific isotopes. It is observed that certain numbers of protons, or neutrons, are "magic" in that an isotope with such a number has unusual stability at the closing of the shell. On the other hand, in certain regions in the chart of the nuclides away from closed shells, nuclei are well described in terms of models of rotating and vibrating states of a deformed nucleus. Development of these models has been dependent upon precise measurements of the nuclear parameters of a large number of specific isotopes.

Such experiments frequently do not require large quantities of the separated isotopes; typical experiments to measure basic nuclear parameters use particle-beam bombardment of either thin target foils or thin deposits of evaporated or plated (milligram to gram) quantities of the separated material. However, these uses are frequently destructive because of losses during preparation of the target foil and damage by the beam. Since advances in knowledge and in techniques of measurement often require repetition of earlier measurements to attain more accuracy, or to search for new phenomenons, the need for separated isotopes for basic nuclear studies is a continuing one.

Some basic measurements in nuclear physics require larger quantities of separated isotopes. Among these are the studies with slow and fast neutrons; with gamma-ray, muon, and pion beams; and with high-energy proton beams. These experiments frequently are not destructive, and the separated-isotope samples may be borrowed from the Isotopes Pool.

The use of separated samples for neutron-cross-section measurements has been very important to advances in basic neutron physics. One significant basic advance to come from neutron physics measurements is the "optical model," a coherent description derived from many neutron (and proton) experiments involving almost all the isotopes. Currently there is promise of developing "unified" reaction theories that combine the descriptions of the shell and the rotational–vibrational structures of nuclei with optical-type theories. The use of separated isotopes has contributed greatly to the arrival at this level of understanding and may be expected to be increasingly important in new progress.

One recent, exciting use of isotopes from the Isotopes Pool has been in the study of muon-capture X rays; this phenomenon offers a completely new tool for investigating the structure of nuclei. Another is in the study of the double beta decay of ^{48}Ca, which requires a large quantity of ^{48}Ca.

The panel has discussed the use of separated isotopes with many laboratory directors at major U. S. university and national laboratories where studies in nuclear structure and intermediate-energy nuclear physics are being conducted. These laboratory directors feel that the small fraction of the operating funds allocated to enriched isotopes is in no way a measure of their importance to their laboratories' efforts and that in most cases these efforts would not even be possible without a continuing supply of the separated isotopes.

The Pake Committee surveyed the progress of physics as a part of the National Academy of Sciences' survey of progress in basic science in 1964. The portion of that report concerning nuclear physics has been updated to 1968 and is to be issued by the National Academy's Subcommittee on Nuclear Structure.* Both these reports, noting the rapid advances in the understanding of basic nuclear physics, repeatedly stress

*Herman Feshbach, Chairman.

both the past importance of and continuing need for separated isotopes in this national program.

Future Needs. The separated-isotopes program has been crucial to the rapid advancement of basic nuclear physics since 1945, and its importance is expected to continue to increase. Nevertheless, the present cost of isotopes for all nuclear physics research is a modest fraction (less than 10%) of the cost of the AEC-supported on-site and off-site programs and a much smaller fraction of the overall national and international research programs that use such samples. During the next 5 years, such costs are likely to become a growing fraction of the operating budgets of university and national laboratories. The growth has been about 10% per year and can reasonably be expected to continue at that rate for the next 5 years.

Applied Nuclear Physics

Three branches of nuclear science may be called "applied nuclear physics": "radiochemistry," or "nuclear chemistry," is principally concerned with fundamental studies of the properties of radioactive nuclei; "shielding physics" is concerned with shielding (man, instruments, etc.) against all penetrating nuclear radiations; and "neutron physics" is concerned with the processes that occur when neutrons penetrate or interact with matter and is the basis of reactor engineering.

Radiochemistry. The nuclear chemists (or radiochemists) have made great progress since 1945. The list of transuranium elements has been extended up to atomic number 105, many isotopes of the ordinary elements have been discovered, and a great deal is known about decay energies, half-lives, etc., of radioisotopes. The applications of this knowledge are just beginning to be appreciated; e.g., the medical doctor, the engineer, and the research scientist, in many instances, can choose, on the basis of this knowledge, a radioactive source to fulfill his particular needs. Frequently the information and the sources themselves would not have been available without separated stable isotopes; in other cases, calutron separation was required after sample preparation by other means—e.g., in a reactor.

One of the principal interests of nuclear chemistry has been the study of fission-produced isotopes. This interest, while not now a major one, may be expected to be important for years to come as radiochemists continue to search for radioisotopes for specific applications in new fields. The space program, as an example, has generated the need for radioisotopes as energy-source fuels in artificial satellites. The separated-isotopes program will continue to be an important aspect of such work.

Of great current interest in radiochemistry are the transuranium elements. The use of separated isotopes has been very helpful in the past in work with these elements, and, with the new accelerators capable of accelerating very heavy nuclei, an increasingly important role may be predicted for separated isotopes. Radiochemistry is a very live field and in the future will no doubt produce many useful radioisotopes. Experiments in radiochemistry will continue to use relatively small quantities (generally less than 1 g) of separated isotopes; nevertheless, because of its broad applicability (industry and medicine), radiochemistry alone could probably justify the isotope separations program, although at a somewhat reduced level of effort.

Shielding, Neutron Physics, and Engineering. Absorption, scattering, and reactions produced by the penetration of nuclear radiations through large masses of matter have become important problems in engineering because of their application to reactor design, reactor shielding, and military and civil defense.

The new science of nuclear physics, with its almost immediate application to engineering, created a problem of communications between the scientists, who make the basic measurements, and the engineers, who need the results of these measurements. As a result, the AEC established two committees—the Nuclear Cross Section Advisory Committee (NCSAC) and the Advisory Committee for Reactor Physics (ACRP)—to coordinate the national needs for applied nuclear physics measurements. These committees publish annually a document, *Request Compilation,* that lists these needs.

Request Compilation forms a useful document for surveying the national needs for basic nuclear measurements. For example, although many of the requests have been satisfied and thus removed from the list, the number of requests increased substantially from 1956 to 1967—i.e., from about 100 to 400—and high-priority requests doubled within the same period. This growth in the number of requests is a good indication of the importance that the applied scientist and engineer attaches to these basic nuclear measurements. About 20% of the requests in the compilation call for use of highly enriched isotopic samples.

Since natural elements are usually the only materials available in sufficient quantity for reactor, shielding, or other construction applications, separated

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isotopes would appear, upon cursory examination, to be of minor importance in this program. However, the measurements requested in the compilation are mostly for use in rather elaborate reactor or shielding calculations, where an analytical expression is frequently much more useful than an empirical curve of cross section vs. energy. A very accurate expression for low-energy cross sections may be found by studying the low-energy resonances of each isotope of an element and then combining these data to express the effective cross section of the natural mixture of the isotopes. This method is practical only when samples of nearly pure isotopes are available. At somewhat higher energies, knowledge of the inelastic-scattering cross section of an isotopically complicated element, e.g., zirconium, is often important. A number of excited states of each of zirconium's five isotopes are involved. The problem is very difficult to solve unless separated isotopes are used to untangle the effects of the many levels involved in the inelastic scattering from natural zirconium. Thus, even when the properties of a natural element are all that is required, the necessary information can be found in some cases only with the help of separated isotopes.

Future Needs. Estimates show that the Isotopes Pool should be at least doubled in size to meet the requirements for large samples for nondestructive use. In particular, present demands and future projections suggest that the isotope-separations operation will need to continue at its present scale for at least 5 more years. At that time another review should be made to compare resources of the pool with requirements for materials needed for cross-section measurements made using new methods now being developed. In addition to the demands on the pool materials, smaller samples of separated isotopes will be needed for destructive uses, for example, for radiochemistry and for measurements where a nuclear explosion is the neutron source.

Nonnuclear Physics

Past and Present Uses. Since most atomic properties are almost independent of the mass of the nucleus, it might seem that there would be little use of separated isotopes in studies of atomic and solid-state properties. On the contrary, very significant contributions to the advancement of knowledge in this field have been made by the use of separated isotopes.

For example, iron isotopes were separated early in the program because of the need to study the nuclear properties of the more abundant iron isotopes for the reactor program and to produce ^{59}Fe [by the ^{58}Fe$(n,\gamma)^{59}$Fe reaction] for blood studies. There was little demand for the ^{57}Fe made available. However, in 1957 Mössbauer discovered that the electron-capture decay of ^{57}Co to a state in ^{57}Fe produced a gamma ray that was emitted and absorbed, without recoil, in suitable solid samples. The use of isotopically enriched ^{57}Fe samples significantly improved these precise Mössbauer studies, and today sales for this purpose are about $40 thousand per year. This history points up an interesting lesson: The store of separated isotopes has an enormous potential value, even when unused, and represents a real national asset that increases in value as science and technology develop.

Another important use of separated isotopes has been in atomic spectroscopy, where the isotope shift due to the hyperfine interaction broadens spectral lines when light from natural elements is measured. To provide sharp wavelength standards, separated-isotope samples of ^{202}Hg are used—e.g., by the U. S. Bureau of Standards.

A foreseeable demand for separated isotopes is in the laser industry, where their use is increasing. An interesting recent application in atomic physics is the use of separated ^{3}He as a gas whose atomic nuclei can be highly polarized by optical pumping. These studies have significantly advanced certain aspects of understanding of plasma and gas-collision physics and have provided an excellent magnetometer for geophysical exploration and military application.

Separated ^{3}He also has played an important role recently in cryogenics: In fundamental studies of matter, ^{3}He cryostats have provided a means for attaining temperatures lower than those available with ^{4}He. Especially important are the ^{3}He–^{4}He dilution refrigerators, which now allow very-low-temperature studies of large samples of matter.

The use of separated isotopes has helped the understanding of phenomenons of isotope effects related to other solid-state research studies—e.g., diffusion in metallurgy, transition temperatures at which superconductivity occurs, and thermal conductivity in matter (understanding of phonon interactions in matter).

The study of defect or impurity centers has been significantly aided with the use of stable isotopes by either employing electron-spin resonance and hyperfine interaction or observing the mass dependence on other physical attributes.

Future Needs. Except for ^{3}He and ^{57}Fe, no large amounts of separated isotopes are at present used in

nonnuclear physics. However, the already large use of these two stable nuclides can be expected to grow in the next few years, and the use of smaller quantities of a large number of isotopes for various basic atomic and solid-state physics research may be expected to continue. The development of unforeseen and unexpected uses of stable isotopes, such as happened with the studies of the Mössbauer effect and with the ^{3}He–^{4}He dilution refrigerators, may reasonably be expected to occur again, perhaps in single-crystal studies of isotopically pure materials for solid-state physics. The recent growth in rate of use of separated isotopes for nonnuclear physics (about 6% per year) may be expected to continue for the next 5 years.

Chemistry

Past and Present Uses. Stable isotopes, used both per se and in the production of radioisotopes, are very significant materials for fundamental research in chemical and chemically related problems. The needs for separated stable isotopes in chemistry lie intermediate between those of nuclear physics and those for programs designed to supply target materials for the production of radioisotopes for tracer methodology in biology and medicine. Chemical research is usually done on a scale that involves only a few grams of material. Radioisotope tracer methodology has become a powerful technique in chemical research, and, in many cases, the tracer production starts with the irradiation or bombardment of a separated stable feed material. The demand for these starting materials will likely continue to grow.

In chemical research the heaviest demand is for enriched isotopes of the light nuclei—^{2}H (D), ^{6}Li, ^{7}Li, ^{11}B, ^{13}C, ^{15}N, ^{17}O, and ^{18}O. The major reason for this emphasis is that many biological and organic processes involve molecules composed mainly of light atoms. The investigation of the structure and reactivity of such molecules can often be facilitated by replacing a high-natural-abundance atom by its less abundant isotopic counterpart.

Infrared Spectroscopy. An important use of deuterium isotopes is in the structural investigation of hydrogenous molecules by infrared spectroscopy. By the substitution of certain of the protons in the molecule with deuterium, significant changes in the fundamental infrared-active frequencies of the molecule can be observed, which, in turn, lead to the identification of the position of the substituent in that molecule.

Nuclear Magnetic Resonance. Since the method of nuclear magnetic resonance (n.m.r.) depends specifically on studies of the isotopes rather than of the element, chemical structure can be deduced by using separated isotopes. Thus deuterated solvents, e.g., CD_3COCD_3, are extremely useful in proton nuclear magnetic-resonance spectroscopy, since the use of these solvents (where chlorinated solvents such as CCl_4 and C_2Cl_6 are unsuitable) results in relatively simple spectrums, which are susceptible to precise measurement and interpretation.

The low natural abundance of ^{13}C makes it difficult to observe n.m.r. signals for ^{13}C in the naturally occurring carbon, and isotopically enriched samples are required. The coupling effects of ^{13}C with either protons or other nuclei that have a magnetic spin moment can be observed by the most advanced techniques. Such measurements, when available, can do much to elucidate the subtleties of molecular structure, and this type of resonance spectroscopy is likely to become increasingly important in chemical research in the next 5 to 10 years. Unfortunately, spectroscopic investigations of this kind are usually quite expensive, since the present cost of ^{13}C is approximately $3 thousand per gram,* and a synthesis procedure, frequently on a scale requiring several hundred milligrams to a gram of ^{13}C, is necessary to prepare the labeled sample.

In a similar context the structure and reactivity of boron compounds have received increasing attention in recent years. Particularly useful in this regard is the investigation of ^{11}B-proton coupling constants by n.m.r. techniques. Although the natural abundance of ^{11}B is approximately 81%, a systematic change in the $^{10}B : {}^{11}B$ ratio makes possible a study of the effect on the n.m.r. spectrum of the substitution of a particular boron atom in a complex molecule.

Reaction Kinetics. Another new technique concerns the study of the isotope effect on the rate of a chemical reaction. It is sometimes possible to follow this isotopic kinetic effect without the use of mass spectrometers for identifying the reaction products, e.g., in the study of the effect of isotopic substitution, using ^{6}Li and ^{7}Li, on the kinetics of electrochemical depositions from solution. Such studies shed light both on the influence of isotope substitution on the kinetic parameters and on the fundamental processes involved in the chemical reaction being studied.

*Prices for ^{13}C of 90% or greater isotopic purity have recently been decreased to $685 per gram by Mound Laboratory, Miamisburg, Ohio.

XUM

Isotopic kinetic-effect studies have also been carried out using ^{15}N (natural abundance 0.37%) rather than the more common nuclide, ^{14}N. However, the present commercially available compounds containing 95% enriched ^{15}N are extremely expensive—e.g., $^{15}NH_3$ costs approximately $1 thousand per gram; consequently the use of this nuclide is somewhat limited at the present time.

Mössbauer Spectroscopy. A major recent development has been the exploitation of the above-mentioned Mössbauer spectroscopy to solve chemical problems. This type of research requires, in some instances, the use of enriched stable isotopes for the production of appropriate sources, in particular, ^{128}Te, ^{118}Sn, and certain rare-earth isotopes. Moreover, the production of the high-specific-activity 250-day isomer of ^{119}Sn requires the availability of highly enriched ^{118}Sn depleted in ^{119}Sn. It is important to note that these significant recent advances in Mössbauer studies were dependent upon the stockpile of separated tin isotopes and parallel the phenomenal advance of Mössbauer work with separated ^{57}Fe. This again emphasizes the separated-isotopes program as an important national asset.

Future Needs. The recent applications of several modern measurement techniques to problems in chemistry discussed above are expected to increase the use of separated isotopes. The modest needs for the heavier elements are expected to grow somewhat, and the demands for the lighter elements are expected to grow substantially. In particular, if the price of ^{13}C and ^{15}N should be reduced, the use of these nuclides would be very greatly increased.

It is clear from what has been said above that the major needs with respect to chemistry and chemical technology lie in the availability of stable isotopes of the lighter elements. In addition, specific stable isotopes of selected elements—primarily ^{57}Fe and ^{119}Sn for Mössbauer-effect studies and ^{13}C and ^{17}O for n.m.r. and infrared studies—are needed. Other possible uses of high-purity separated stable nuclides are as standards for mass spectrometry and chemical spectroscopy (e.g., ^{40}Ca completely free of strontium and barium).

Although it is difficult to establish the particular stable isotopes that will be of greatest importance in terms of fundamental research in chemistry in the near future, it is probably valid to assume that this country should have the capability of providing stable isotopes of all elements for continuance of active research in chemistry.

Earth Sciences

Past and Present Uses. Although sales of separated stable isotopes for use in earth sciences account for only about 1% of the output of the program, the availability of separated stable isotopes has profoundly influenced the course of earth sciences over the past two decades. The primary benefits have resulted from stable-isotope-dilution techniques, which permit elements having more than one isotope to be determined reliably within accuracies of a percent or less at the microgram level. The foremost application has been in geochronology. Since 1948 the concentrations of radioactive parents (U, Th, Rb, and K) as well as of radiogenic daughter elements (Pb, Sr, Ar, and Ca) have been determined largely by stable-isotope-dilution techniques. Approximately 200 U. S. publications on geochronology describing these methods can now be cited.

Besides geochronology, isotope dilution has been useful, along with neutron activation, in measuring concentrations of trace elements in rocks and meteorites. In some cases serious errors in cosmic-abundance data were first discovered by stable-isotope-dilution techniques.

Still another application is the determination of absolute abundance data for isotopes of some elements. If an element has four or more isotopes and at least two of them can be obtained in high purity, absolute abundance data can be obtained. These methods have been used to show that reported variations in abundances were in some cases due to isotopic fractionations introduced in the chemical processing or in the mass spectroscopy. The method is also being used to reliably establish small variations in isotopic abundances in lead samples, and the results show promise for the development of an isotopic tracer for studies of geological processes.

The value of the stable-isotope inventory in the earth sciences has increased as the number of separated isotopes has increased and especially as the isotopic purity of available isotopes has increased. None of the experiments cited above consumes a large amount of material—usually 1 to 50 μg of a separated isotope per analysis. Purchases by laboratories are generally in the 10- to 100-mg range for any particular isotope, and the total yearly consumption of separated isotopes at present is estimated to be 150 mg.

Future Needs. As far as can be seen, future demands for separated isotopes will be similar to, although somewhat larger than, those of the past. The reasons for the larger demand are the increases in both

the number of workers and the number of applications of the techniques. The number of workers in isotope geology is now increasing at a rate of approximately 10% per year, and the consumption of separated isotopes in earth-science research can be expected to increase by a like amount for that reason alone. New isotope laboratories have heavy initial requirements for separated isotopes, but it is difficult to predict the increase in consumption resulting from new techniques; e.g., the relative abundance of rare earths in rocks, a useful parameter in the study of problems in petrogenesis, until recently has been determined by neutron activation. However, in the past 2 years, two laboratories have started to make the determinations by isotope dilution, creating a new demand for separated isotopes of some 10 rare-earth elements.

Still another factor increasing the demand for separated isotopes will be the availability, and required analysis, of lunar samples expected in the next few years. More than 50 different laboratories are now developing methods for analyzing lunar material with high precision and low sample consumption, and more than a fourth of these schemes will involve separated isotopes. Samples of enriched isotopes of many elements will be needed.

Although earth-science research has not been a major consumer of separated isotopes, its progress in the past has been tied to the stable-isotopes program in many important ways. This association will certainly continue to exist in the future.

Biology and Medicine

Stable isotopes are needed in quantity to provide the target material for reactor or cyclotron activation in the production of radioisotopes, which are rapidly increasing in importance as tools for diagnosis and therapy. For diagnosis, common requirements are a short half-life to minimize the radiation exposure to the patient and emission of a gamma ray to permit detection of the isotope from outside the body. Isotopes of all elements are possibly useful, as most elements can be attached to other molecules or particles. Radioisotopes are also useful for therapy when selective concentration (in the body) can be achieved. The requirements in this application are primarily for beta emitters of moderate half-life.

Diagnosis. Medical diagnosis involves both scanning and biochemical measurements.

Scanning Procedures. Radioisotope scanning to locate tumors and determine other tissue abnormalities has become a routine clinical procedure. Many types of localizing agents have been used, but the ever-present questions are: Which agents are most effective? How can they be compared objectively? And what are the biological and biochemical factors for establishing a compound's usefulness in scanning? This last question is important in understanding how scanning agents work and in placing this diagnostic procedure on a more rational and less empirical basis. In the past, compounds have been selected for such use primarily on the basis of the nuclear properties of the isotope and only secondarily for biological reasons.

With the development of scintillation cameras and high-speed scanners, the use of very-short-lived isotopes in large doses has become an attainable objective. Large amounts of specific tracer activity allow (1) more rapid procurement of the diagnostic picture, (2) lower radiation dose to the patient than is obtained with isotopes of longer half-life, and (3) utilization of the increased information content for finer picture resolution.

Technetium-99*m* is an increasingly popular radionuclide label for emission scanning because it has an optimum (140 keV) photon energy; a short (6 hr) half-life; no energetic beta emission; and favorable chemical properties. The medical use of ^{99m}Tc has resulted in a great demand for its parent nuclide, ^{98}Mo; 10% of 1966 target-material sales were enriched ^{98}Mo. Studies with ^{47}Ca and ^{85}Sr have shown that scanning for bone diseases may yield positive results despite negative radiographic attempts to demonstrate such diseases. An enumeration of each of the specific organ systems in which radioscanning has been effectively used includes all the organ systems in man.

Among the scanning isotopes that are prepared from electromagnetically separated stable isotopes (given in parentheses) are ^{99m}Tc (^{98}Mo), ^{74}As (^{74}Ge), ^{64}Cu (^{63}Cu), ^{197}Hg (^{196}Hg), ^{203}Hg (^{202}Hg), ^{113m}In (^{112}Sn), ^{47}Ca (^{46}Ca), ^{85}Sr (^{84}Sr), and ^{131}I (^{130}Te). In addition to these nuclides with relatively short half-lives, radionuclides with ultrashort half-lives are gaining wider use, including several metastable isotopes that are prepared from stable isotopes available from the ORNL's Isotopes Division—5-sec ^{191m}Ir (^{190}Os), 40-sec ^{109m}Ag (^{108}Cd), and 13-sec ^{81m}Kr (^{79}Br).

An estimate of the importance of the stable-isotopes program to medical diagnostic procedures can be obtained from an examination of the isotope sales in 1966 and 1967. One-third to one-half of the sales are for isotopes that are the precursors of the radionuclides used in medical diagnosis. Two hundred hospitals are now equipped for scanning, as compared to

XUM

none in 1963. For example, Johns Hopkins Hospital performs 6 to 7 thousand scans per year, each using 10 to 15 mc of ^{99m}Tc. Many of the most important of these precursor isotopes are in the group that would be exhausted within 2 years were the separations program to be discontinued.

Nuclear medicine is still in its infancy and is just now beginning to make use of materials and techniques that have heretofore remained in the domain of the physicist. If the accelerated progress of nuclear medicine in the past 5 years is projected into the future, it may be anticipated that the development of new techniques will increase the demands for separated isotopes. While, in some instances, nuclides of low enrichment may be adequate, the availability of highly enriched nuclides as targets will reduce the dangers from the administration of toxic substances as well as from undesirable radioisotopes of the same element.

Biochemical Measurements. Although a very large proportion of radioisotopic diagnosis is concerned with scanning, an extremely important and rapidly developing technique in medical diagnosis is the measurement of organ functions with radiotracers. Crude estimates and laborious chemical procedures have now given way to accurate and simplified radioisotope methods for the evaluation of many physiological processes.

Studies of thyroid function were among the first in which radioisotopes were used. While the commonest iodine radioisotope (^{131}I) used in those studies is generally produced by fission or by the use of normal-tellurium targets, it is also produced from enriched ^{130}Te. Extensive work on iron absorption and incorporation into red blood cells in cases of anemia has been made possible by the availability of two iron radioisotopes produced from enriched ^{54}Fe and ^{58}Fe obtained from the ORNL Isotopes Division. The radioisotopic diagnosis of pernicious anemia has become a routine procedure with the availability of radionuclides of cobalt, several of which are transmutations of enriched stable isotopes. Various measurements on the circulatory system, of critical importance in the diagnosis of myocardial abnormalities, are now made with radioactive tracers. While the isotopes of rare gases are being used increasingly in these studies, the isotopes of rubidium, iodine, and chromium, prepared from enriched stable nuclides, do play a role.

An abnormal pulmonary ventilation–perfusion relation is considered to be an important cause of hypoxemia. The effects of this condition could be studied with separated stable ^{17}O and ^{18}O if these isotopes were available in large quantities at moderate costs. Measurements of blood volume, total body water, and electrolytes with a multitude of radioactive isotopes have become routine in the evaluation of depletion syndromes, e.g., severe burns. Calcium and copper nuclides have been extensively used to provide radioactive tracers for studies on absorption and metabolism of these elements. Kinetic studies with ^{85}Sr [from the ^{84}Sr$(n,\gamma)^{85}$Sr reaction] have been successfully used in the evaluation of hypoparathyroidism. Selenomethionine labeled with ^{75}Se [from enriched ^{74}Se$(n,\gamma)^{75}$Se] has also proved of value in parathyroid and pancreas studies. Malabsorption syndromes, exudative enteropathies, and liver- and kidney-function and structure studies are among the areas in which radioisotopes are being extensively used. The life-span of blood cells as well as fecal blood loss may be routinely studied by tagging of cells with ^{51}Cr [from the enriched ^{50}Cr$(n,\gamma)^{51}$Cr reaction].

With increasing capabilities in medicine and surgery, the need for rapid evaluation of structure and function of organ systems is increasing. Separated stable isotopes as well as radioisotopes, many of which are produced from separated-stable-nuclide targets, are essential for this evaluation. Laborious, costly, and cumbersome chemical procedures are gradually giving way to the more precise, inexpensive, and simple analyses made possible by the use of isotopes.

Activation Analysis. Although all types of activation analysis may well play a future role, present medical emphasis is on the well-developed technique of high-flux-neutron activation analysis. Medical applications of this technique are still in their infancy but are already appreciable. Activation analysis provides a nuclear method for the quantitative determination of the elements in samples, regardless of the chemical states of these elements. The application of activation analysis to the detection of trace elements offers the merits of tracer techniques without any theoretical or real danger of exposure to radiation.

The usefulness of stable isotopes for activation analysis depends on a low natural isotopic abundance, availability of an inexpensive enriched form, an adequately large neutron cross section, and an easily detected and readily determined product. In pilot studies of metabolism, trace-element assimilation, etc., activation analysis, preceded by feeding of specific stable nuclides, has been found to be a simpler and more accurate method than previous chemical procedures for determining small amounts of materials. A revolution in the clinical chemistry of medical diag-

nostic centers may well be the result of these initial studies.

While the use of stable nuclides for these studies is still small, the availability of these nuclides is essential for evaluation of the methods. It is not unrealistic to expect a substantial increase in the needs of this developing program for separated nuclides.

Therapy with Isotopes. Radioisotopes, which are well known, and stable isotopes, to a much lesser extent, have been used for therapy.

Radioisotopes. Radioisotopes are used extensively in the therapy of malignancy as well as in the treatment of physiological abnormalities of the thyroid gland. Very few of these radioisotopes, however, are prepared from stable isotopes. As mentioned previously, although ^{131}I used in the diagnosis and treatment of thyroid diseases may be prepared from enriched ^{130}Te, it is more often made from normal Te or is of fission-product origin. At present the only radioisotopes used in therapy for which ORNL provides target materials are ^{85}Sr, used in treating bone cancer; ^{51}Cr (from ^{50}Cr) and ^{192}Ir (from ^{191}Ir), which are implanted in tumor-bearing areas; and ^{82}Br (from ^{81}Br) used in the treatment of patients with bladder tumors.

This short list includes only the radioisotopes that have been or are currently in clinical use. A list of the radionuclides being experimentally tested would include a very large proportion of the radioactive isotopes available and some not so readily available. As long as clinical medicine is faced with the largely empirical evaluation of each radiation source, the necessity for providing a variety of radiation sources remains.

Stable Isotopes. Stable nuclides that have large neutron cross sections, such as ^{10}B and ^{6}Li, have been used in therapy experiments. When these nuclides are bombarded with thermal neutrons, alpha particles are produced by the reactions $^{10}B(n,\alpha)^{7}Li$ or $^{6}Li(n,\alpha)^{3}He$. These alpha particles have an extremely high ionization efficiency and a very short range within tissue, and thus they expend nearly all their energy within the irradiated tumor. As additional information on stable nuclides becomes available, their use for therapeutic purposes can be expected to grow.

While progress in the therapy of malignancy has been slow, radiotherapy has been and continues to be an integral part of cancer treatment. Experiences with stable nuclides of large cross section suggest the possible usefulness of certain nuclides as an adjunct to cancer therapy. The lack of availability of a variety of such nuclides could be disastrous to developmental research in assessing the possible clinical usefulness of such an approach. Great advances in medical scanning have been made because of the availability of the separated nuclides, but the choice of specific nuclides has been determined largely on an empirical basis. A similar area of empirical examination, i.e., the usefulness of neutron-capture therapy, appears to be in the immediate offing.

Biology. Separated stable isotopes are used in biology, both directly and as target materials for the preparation of radioisotopes.

Direct Uses. Biological experiments are generally concerned with the major constituents H, C, O, N, P, and S, and to a lesser extent with the ions of Na, K, Mg, Ca, and Cl. Direct use of stable isotopes has been extremely limited because radioactive isotopes of most of these elements are more convenient to use, easier to detect, and cost less. Although oxygen and nitrogen do not have convenient, readily available radioisotopes, most oxygen and nitrogen pathways can be traced indirectly by molecules containing ^{3}H or ^{14}C. Technical improvements in n.m.r. and mass spectrometry, however, now make it possible to study oxygen and nitrogen directly. Oxygen-18 has been used in studies on biologically important organic compounds. Identification of ^{17}O by n.m.r. and identification of the other stable oxygen nuclides by mass spectrometry have enhanced our knowledge of the metabolism of oxygen-containing compounds. Nitrogen-15 has been used primarily as a density label, but it has also found usefulness in determining the mode of action of such nitrogen-containing compounds as phenylhydrazine. Although experiments basic to the understanding of the mode of replication of nucleic acid were carried out with ^{15}N, the present use has become quite limited. The primary reason ^{15}N has not gained wider use as a density label in animal-cell experiments is the very high cost of ^{15}N-containing compounds needed for adequate density alterations. If one were able to adequately density-label viruses, for instance, the detection of the invading nucleic acid in the host cell would be a relatively simple matter. Carbon-13, like ^{17}O, can be detected by either n.m.r. or mass spectrometry. Its current limited usefulness in organic syntheses and molecular-arrangement determinations will undoubtedly be expanded with continued technical improvements. The availability of inexpensive infrared spectroscopes capable of detecting an excess of several tenths of an atom percent of D in body

XUM

water has greatly widened the scope of investigations with D. In recent studies of fatty-acid synthesis, a specific metabolic defect has been demonstrated by maintaining a 0.5 at.% excess of D in a patient over a period of 5 months.

The minor elements (Fe, Mn, Cu, Co, I, etc.) are generally present in such small quantities in biological systems that radioactive tracers are required for their in-situ study. The demonstration of the possible biological applications of the Mössbauer absorbers ^{57}Fe and ^{119}Sn, however, has yet to be fully evaluated.

Target Materials. The primary biological use of enriched stable nuclides is in the production of high-specific-activity radioisotopes. With the exceptions noted above, most of the biologically important elements have useful isotopes. Although ^{32}P is by far the most important, progressively increasing use is being made of radioisotopes of the trace elements, e.g., ^{64}Cu, ^{45}Ca, ^{47}Ca, and ^{85}Sr.

Ion fluxes following electrical excitation of cell membranes are frequently measured by determining the movement of ^{24}Na and ^{42}K. The role of specific trace elements in plant and animal growth and maintenance studies has been determined to a very large extent by the use of a variety of readily obtainable radioisotopes. It is not possible to assess the importance of this availability to the great mass of biological information that has been generated.

Projected Medical and Biological Needs. Major technological advances in mass spectrometry, n.m.r., and infrared spectroscopy have brought about a renewed interest in low-abundance isotopes: D, ^{13}C, ^{15}N, ^{17}O, and ^{18}O. For useful studies in animal cells, these isotopes must generally first be incorporated into costly synthetic precursors; the initial cost of these nuclides is also a primary deterrent to their use. A major reduction in the cost of the stable isotopes would almost certainly be translated into a very great demand for increased quantities of materials containing them. Great strides have been made in n.m.r. methods for determining ^{13}C and ^{17}O compounds, and such methods are now being evaluated for their potential medical and biological usefulness. If these compounds are made more readily available by substantial reductions in cost, we may anticipate an exponential increase in their utilization.

The major medical and biological use of stable isotopes is as precursors in the production of radioisotopes. From a comparison of the sales of stable isotopes destined to be targets for radioisotope production with the total sales of stable isotopes, we may conclude that 10 to 12% of total calutron operating time is now devoted to the separation of isotopes important in medicine and biology. Medicine, in particular, is in a phase of accelerating use of these isotopes. If the present rate of increase in demand for the single isotope ^{99m}Tc should continue and be given a sufficiently high priority, all available calutrons would have to be turned over to production of ^{98}Mo. Most medical facilities are only beginning to make use of the diagnostic potential of ^{99m}Tc. Obviously, this accelerated demand is not likely to continue. But what of other developments? Almost certainly additional isotopes with great promise in diagnosis and therapy will be found in the very near future. It is conceivable that, before too long, all the present calutron potential will be required for medical uses. Although such projections may be fraught with dangers, the dangers of more conservative predictions to the development of nuclear medicine and to contributions to the nation's health are far more serious.

Administration of Programs

The AEC stable-isotope separations programs at ORNL and Mound Laboratory are conducted in extremely close collaboration with the users of the products and are modified from time to time. The Mound program, narrower in scope and smaller in size than that of ORNL, is essentially self-supporting from sales. The ORNL program, because of its responsibility to supply separated materials to the loan pool, is not self-supporting.

Conclusions

Summary of Needs

Basic Nuclear Physics. The use of separated stable isotopes as targets for beam bombardment in the study of the basic properties of nuclei is of fundamental importance. It is clear that the rapid advances made in understanding nuclear forces and nuclear structure could not have occurred without the AEC stable-isotope separations program. Nuclear studies started out using nearly all the products that the program produced and have since then used an increasing quantity, although decreasing fraction. The AEC-supported nuclear laboratories account for just under half the sales, yet these costs are only a few percent of

these laboratories' operating budgets. Curtailment of this supply would effectively destroy nuclear research.

Applied Nuclear Physics. The applied nuclear physics programs—radiochemistry and neutron and reactor physics—are very dependent in their advancement on the purchase of separated isotopes and on use of isotopes from the Isotopes Pool. In turn, the reactor-development programs and many other applied-research programs are strongly dependent upon the separations program. Goals set in 1957 for the pool have not actually been met, and it is now evident that the newer methods of cross-section measurement require that the size of samples in the pool be increased to about $^{2}/_{3}$ gram atom each for important isotopes.

Nonnuclear Physics. While the scale of use of separated stable isotopes in nonnuclear physics has been modest, the contributions of the AEC separations program to nonnuclear physics has been, in many cases, dramatic and of fundamental importance. If some areas of gas-, liquid-, and solid-state physics are to continue to advance, the separations program is essential.

Chemistry. Nonnuclear physics and chemistry together account for about a fifth of the separated isotopes sold. When coupled with modern measuring methods, the use of separated isotopes has allowed basic new advances of understanding of chemical structure, reactions, and kinetics. The need for separated stable isotopes in chemistry is expected to grow, and there is an especially obvious growing need for the separated isotopes ^{13}C, ^{15}N, ^{17}O, and ^{18}O.

Earth Sciences. A supply of separated stable isotopes has been of great importance to advances in geochronology and the determination of the cosmic abundances of the elements. While there is little need for separated isotopes in large quantity, a continuing supply of isotopes of many elements is crucial for continued progress.

Biology. Biological research, being mainly concerned with compounds of light elements, has found little use for stable nuclides of heavier elements. As in chemistry, stable nuclides of carbon, oxygen, and nitrogen are potentially very useful, but commercial production of these isotopes has not yet resulted in low-cost products. Consequently, biology laboratories have not installed the modern instruments needed to utilize stable isotopes. The AEC production of these isotopes at low cost might bring much greater use and allow major advances in biological studies.

Medicine. Medicine and biology combined use just under half the separated stable isotopes produced. At the inception of the stable-isotope program, no one could have envisioned the demands medicine would make on this program. From the token use of stable nuclides 10 years ago, medicine and its allied disciplines have extended the application of these nuclides to the point that one-third to one-half of present sales are for biological and medical studies. Despite the recent phenomenal growth of medical usage of isotopes, only a relatively small number of medical facilities are employing the diagnostic and therapeutic capabilities provided by isotopes. The full realization of the general applicability of radioisotopes is likely to continue the exponential rise in the demand for the precursor stable nuclides. The increased application, as well as the continued exploration, of medical uses of isotopes may well require the expansion of present stable-isotope separations facilities. At present growth rates, medical uses along with other uses of separated isotopes will overtax the entire present capacity of the separation facilities in a few years. The curtailment of the present stable-isotopes program, or the failure to plan now for expansion for the future needs of medical users of isotopes, would be disastrous to the development of nuclear medicine in the United States.

New Needs. While it is dangerous to predict the future, the Committee strongly believes that our society is entering an era of nuclear and quantum technology that will be different from that of the present, which is based largely on nonquantum, nonnuclear science. The uses of separated isotopes in this new technology—beyond those envisioned and discussed above—will probably be multifold. Among the diverse uses that may become important in the future are structural components of nuclear reactors and devices, industrial product labeling, measuring and controlling air pollution, and industrial process and product control.

State of Inventory

Two-hundred thirty-four different isotopes were listed for sale on the Jan. 31, 1968, inventory of stable isotopes. Of these, 81 are available in a supply of more than 1 gram atom of 90% or greater isotopic purity. Forty of them are not available in isotopic purities as great as 90%. Some of the stable isotopes that are used as primary sources for production of radioisotopes are being sold at a rate greater than 1 gram atom/year. The sales of ^{98}Mo involved more than 5 gram atoms in FY 1968! While an inventory of 1 gram atom of

XUM

high-purity material may be adequate for nuclides that are not currently used for radioisotope production, it is obvious that a limited view of production needs could be disastrous to the development of such procedures as medical radioisotope scanning. If one examines the increased demand for ^{98}Mo over the past several years, one is impressed by the fruitlessness of estimating the "numbers of years of supply" of a given isotope. The demand for ^{98}Mo has increased approximately sevenfold since 1966; ^{57}Fe has shown a similar increase. Despite the limitation of an analysis based on the years of supply available of a given isotope, such a calculation was made, assuming demand at the 1967 level. These numbers imply a maximum number of years of supply of:

Maximum years of supply	Number of isotopes
0 to 2	24
2 to 5	26
5 to 10	26
10 to 20	30
20 to 50	55
50 to 100	36
>100	28

Thus the supply of 24 isotopes would be exhausted at the end of a 2-year shutdown, including half of those used for the production of radioactive tracers.

State of the Isotopes Pool

The total number of gram atoms proposed as the objective of the pool when set up in 1957 to be available for loan for nondestructive use has been exceeded; yet, three-eighths of the isotopes are in quantities below the old objective, and a number have been overproduced because of their high natural abundances. The goal, therefore, actually has not been attained. In addition, new measurement techniques will require larger samples than those envisioned in 1957 and will require that the quantities listed in the 1957 objectives be approximately doubled. Finally, the need for ultrahigh isotopic purity of some nuclides requires multiple-pass separations and indicates that the separations effort will need to be continued.

International Trade

Approximately 40% of the total sales are for nuclides used by commercial enterprises for production of radioisotopes. If the stable nuclides were not available in the United States, these precursor nuclides would almost certainly be purchased from foreign sources, or the radionuclide itself would be purchased from readily available foreign sources. At present, approximately one-third of the sales of stable nuclides is to foreign countries. The total annual income from sales of stable isotopes for radioisotope production is about $500 thousand. If foreign sources were to supply domestic demands for either the stable precursors and/or radioactive products, the net dollar outflow would be far in excess of this figure. Under present arrangements the stable-isotope separations program provides a source of foreign currency as well as discouraging purchases of nuclides abroad.

Recommendations

Primary Support

The Division of Research of the AEC should continue to provide primary support for the entire program so that production will not be directly and immediately controlled by current sales. While responsibility for the program rests with the Division of Research, an effort should be made to make the program more self-sustaining.

To endanger the program by making its continued operation contingent upon self-support would, at this time, seem extremely unwise. There is general agreement, however, that the future development of the program should take into account the desirability of reducing the unreimbursed support from the AEC. Apart from the scientific justification for the continued direct support by the AEC, this Committee believes that the entire program must be viewed as a national resource of incalculable value to the United States. While it required less than a million dollars to rehabilitate the calutrons now in use in the separated-isotopes program, the actual cost of these separators (built during the war for electromagnetic separation of the uranium isotopes) was about $\$50 \times 10^6$, and their replacement now, no doubt, would cost more than $\$100 \times 10^6$. The opportunity to apply so inexpensively such machines for science will never occur again. British and French stable-isotope separations programs have not been very successful economically because of the lack of such a war-born program in those countries. The calutron separators are assets to science and should on no account be allowed to fall victims of disuse, disrepair, or cannibalism. The now unused facilities should be preserved for future expansion.

Inventory Reevaluation

The present inventory should be reevaluated to reflect replacement cost at current operating efficiencies, not the accumulated cost of the program. Isotopes of low enrichment should be reduced in value in proportion to reduced demand. Where an inventory nuclide of lower enrichment has been replaced by a more highly enriched nuclide, the nuclide of lower enrichment should be depreciated to its asset value as an enriched feed material for further calutron enrichment.

Revision of Nondestructive Loan Program

Over the past 3 years, the monetary value of material on loan has increased to a level of about $\$5 \times 10^6$ per year. Since at least half the calutron time is spent on the production of nuclides designated for the loan pool, it appears appropriate that a share of the cost of the operation of the facility might be borne by the loanees, and the committee has considered this possibility. In addition to distributing the cost of operation of the facility to those who conduct research that is nondestructive with regard to the isotopes, a rental fee would help eliminate hoarding of pool nuclides. While any charges will of necessity be somewhat arbitrary, the goal of such charges might be to achieve a revenue of \$1 to 1.5×10^6 annually. This would require imposing an annual charge of approximately 10% of the current inventory value and would undoubtedly impose a major financial burden on many universities. An even more serious effect would be the discouragement of experimenters who might otherwise wish to make a much needed measurement. For this reason the Panel does not recommend that a charge be made for loans from the pool. Nevertheless, some revision of the loan program must be made to prevent the holding of nuclides for unreasonably long periods. A compromise scheme that would assess the borrower—perhaps 1% of the current inventory value for an initial 5-month period followed by an increased charge of, say, 0.5% per month—may be acceptable. Under no circumstance should any nuclide be loaned to an individual for a continuous period of more than 24 months without a specific review of the work in which it is being used.

Revision of Pricing Policies

[Editor's Note: Sales isotopes are priced on a full recovery basis (which includes amortization and the AEC 15% overhead fee), with income going directly to the U. S. Treasury.]

Relative prices should be based on production costs with charges allocated among isotopes according to demand. Ideally, one would multiply all prices by an appropriate factor so that the previous year's sales plus rentals would recover the coming year's budget. Based on the previous year's sales, such a policy would result in a two-fold increase in the price of isotopes. After carefully considering such a major price increase for isotope consumers, the committee has rejected this proposal. While such a policy would relieve the AEC Division of Research from subsidizing the program, the net effect would be to increase federal expenditures in other agencies. For example, a doubling of the price of ^{196}Hg or ^{98}Mo would be immediately translated by commercial sources into an increased cost to medical scanning facilities. Since a large share of the cost of maintaining such facilities is ultimately borne by other federal agencies, these price rises would be translated into increased costs for other agencies such as Health, Education, and Welfare. The budget for stable-isotope separations at ORNL would be balanced at the expense of other agencies—an expense far greater than is presently being subsidized by the AEC Division of Research. A two-fold increase in the price of the stable nuclides may not be the ultimate answer to the problem of supporting the stable-isotopes program, but some price adjustment seems justified. Increases in price to small consumers will probably have little or no effect on a large proportion of the users. Increases in prices to commercial sources, however, will almost certainly be translated into further increases in the ultimate federal expenditure for radionuclides.

Program Emphasis

The scientific and technical arguments given above indicate that a new and significant emphasis should be placed on the following aspects of the program:

1. Increasing capabilities of producing materials of high isotopic purity by improved techniques and by multiple-pass separations.
2. Expanding AEC efforts in the separation of light elements (especially those with biological and medical importance) if larger scale, less expensive production methods can be developed.
3. Studying ways to expand the capabilities of the present separations program to be better able to handle foreseeable rapidly growing needs—especially in medical applications. (REG, PSB)

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Chemistry of the Electromagnetically Separated Stable Isotopes

Abstract: *The purification method for the stable isotopes of tungsten electromagnetically enriched at Oak Ridge National Laboratory is described. The tungsten oxide from the ignition of the graphite collection pockets is dissolved in ammonium hydroxide and then precipitated as tungstic acid with concentrated nitric acid. Residual tungsten in the filtrate is recovered by precipitation with cinchonine. The combined precipitates are ignited to WO_3, the stable inventory form.*

This is the fourth article of a series[1-3] on the chemistry of electromagnetically separated stable isotopes enriched at Oak Ridge National Laboratory. General problems and methods associated with the handling and purification of isotopically enriched materials and with their chemical and isotopic analysis were described in the first of the articles. This first article also included a description of the chemical processing methods for enriched isotopes of Sr, Ba, Pb, Zr, and Hf. The second article described the processing of Cr, Pt, and Os isotopes and the third the isotopes of cerium. The description of the recovery and purification of separated tungsten isotopes follows.

Tungsten

The element tungsten has five naturally occurring stable isotopes. The method (Fig. 1) for chemical purification and recovery of the tungsten isotopes is relatively simple: the bulk of the tungsten isotope is precipitated as tungstic acid, and the residual tungsten in the filtrate is recovered by precipitation with cinchonine.

The most recent separation of tungsten isotopes[4] was in response to a request by the National Aeronautics and Space Administration (NASA) for 5 lb of ^{184}W of greater than 90% isotopic purity together with all other tungsten isotopes that could be collected concurrently. The results of this recovery effort (Tungsten Series MC) are

Isotope	Natural abundance, at. %	Abundance after enrichment, at. %	Recovery, g
^{180}W	0.135	~6.5	37
^{182}W	26.41	~94	1957
^{183}W	14.40	~75	23.7
^{184}W	30.64	~94	2345.6
^{186}W	28.41	~97	1866

The isotopes of tungsten are collected in graphite pockets. The outside surfaces of the pocket are carefully cleaned by buffing with Al_2O_3-abrasive cloth, and then the thick, heavy ends are removed to within $\frac{1}{8}$ in. of the body of the pocket with a hacksaw (removal of the ends reduces ignition time and lowers the quantity of impurities, with essentially no loss of the collected isotopes). The tape covering the opening in the pocket is removed, and the pocket is crushed in a plastic sack or in plastic sheet wrappings.

The graphite pieces are transferred to a quartz boat; then the boat and contents are ignited at 750°C in oxygen in a horizontal Vycor tube, and the oxidation is continued until all the graphite is consumed, at which time the tungsten is present as WO_3. This ignition temperature is a compromise based on literature references which state that WO_3 is volatile above 750°C (Ref. 5), at ~850°C (Ref. 6), above 800°C in vacuo,[7] or above 900°C (Ref. 8). The validity of the 750°C ignition temperature was tested by igniting eight ^{186}W collector pockets at 750°C, bubbling the off-gas through a water trap, washing the ignition tube with concentrated NH_4OH, and adding the wash solution to the trap solution for the subsequent determination of tungsten. No tungsten was found.

The WO_3-containing residue is transferred in 25-g lots to 1-liter plastic beakers, and approximately 750 ml of NH_4OH (free of SiO_2) is added to each 25-g sample. Each beaker is covered with a plastic lid held in place with a lead weight and is then immersed in a steam bath to digest the solution; digestion is continued until approximately 250 ml of the solution evaporates.

The beaker is removed from the steam bath and refilled to the original level with concentrated NH_4OH; the digestion on the steam bath is continued. Usually two additions of NH_4OH and a digestion period of 8 hr will be required to effect solution* of the WO_3.

The ammoniacal solution of ammonium tungstate is filtered in plastic equipment to avoid silica contamination, and the filtrate is added, while stirring, to approximately 250 ml of hot (near boiling) concentrated HNO_3 to precipitate yellow $H_2WO_4 \cdot H_2O$ (or

*The procedure for recovery of small amounts of insoluble residue that sometimes remains is given in Ref. 4.

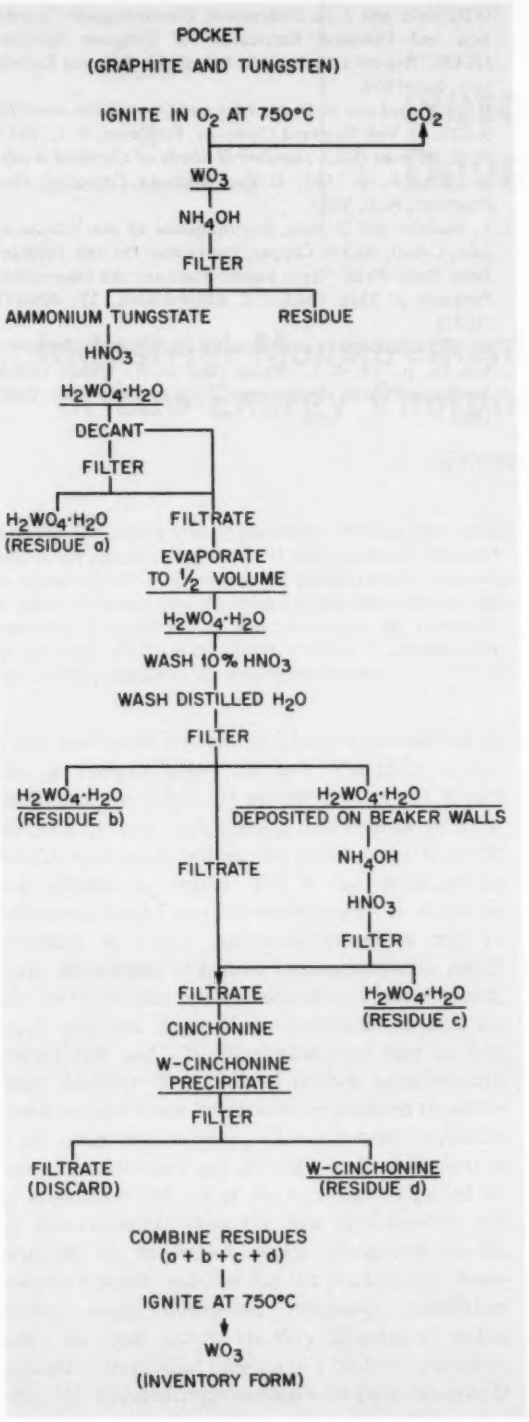

Fig. 1 Purification method for enriched isotopes of tungsten.

$WO_3 \cdot 2H_2O$), which is slightly soluble in HNO_3 and is difficult to filter and wash.

The mixture of solution and precipitate is digested on a steam bath for 20 to 30 min to flocculate the finer particles (the color usually changes from light to dark yellow during digestion). The mixture is cooled, the supernatant liquid is decanted, and the tungstic acid residue is washed onto Whatman No. 42 filter paper with 10 vol.% HNO_3. Any precipitate remaining in solution can be recovered by evaporating the combined solutions (decanted liquid and filtrate) to approximately one-half the original volume on a steam bath. The tungstic acid crystallizes, and most of it deposits on the sides and bottom of the beaker during evaporation. After evaporation, the contents of the beaker are filtered to separate any loose $H_2WO_4 \cdot H_2O$ residue, and both the deposited tungstic acid and the loose residue are washed with 10% HNO_3, then with distilled water. The $H_2WO_4 \cdot H_2O$ still adhering to the beaker walls is dissolved in concentrated NH_4OH and again is precipitated with hot HNO_3. The mixture is filtered on paper, and approximately 5 ml of cinchonine solution (prepared by dissolving approximately 125 g of cinchonine in 1 liter of 1:1 HCl solution) is added to the combined acid filtrates and wash solutions from the above-mentioned steps to complex and precipitate any dissolved tungsten. This mixture is heated to just below boiling for at least 30 min and subsequently cooled to allow the precipitate to settle. Some ashless paper pulp is added, and the precipitate is transferred to a small filter paper using a hot cinchonine wash solution.[6]

All filter papers are collected—those containing $H_2WO_4 \cdot H_2O$ residues and those containing tungsten–cinchonine precipitate—and the combined residues are ignited in a Vycor dish at 750°C to WO_3, the preferred inventory form.

Although attempts are made to prevent SiO_2 contamination of tungsten by using plastic instead of glass equipment for the ammoniacal solutions, spectrographic analysis of the purified WO_3 occasionally shows silica as an impurity.

If the level of the silica impurity is greater than 0.5%, the silica is removed by the following procedure: The WO_3 is transferred from the Vycor dish to a platinum dish and moistened first with concentrated H_2SO_4, then with HF (approximately 10 ml of each reagent); the moistened residue is heated to complete dryness on a hot plate in a fume hood; and the dry residue is transferred to an electric muffle furnace and heated for 1 to 2 hr at 750°C to drive off the silicon as

XUM

SiF_4 (the H_2SO_4 is added to prevent volatilization of tungsten). The procedure is repeated if necessary.

(REG)

REFERENCES

1. R. E. Greene and P. S. Baker, The Chemistry of Electromagnetically Separated Stable Isotopes, *Isotop. Radiat. Technol.*, 3(3): 193-200, 203-204 (Spring 1966).
2. R. E. Greene, The Chemistry of Electromagnetically Separated Isotopes, *Isotop. Radiat. Technol.*, 3(4): 312-317 (Summer 1966).
3. R. E. Greene, The Chemistry of Electromagnetically Separated Stable Isotopes, *Isotop. Radiat. Technol.*, 5(2): 101-103 (Winter 1967–1968).
4. L. O. Love, K. A. Spainhour, W. A. Bell, W. C. Davis, R. L. Johnson, J. G. Tracy, C. W. Sheridan, A. H. Snyder, F. R. O'Donnell, and J. N. Underwood, Electromagnetic Separation and Chemical Refinement of Tungsten Isotopes, USAEC Report ORNL-4405, Oak Ridge National Laboratory, July 1969.
5. H. H. Willard and H. Diehl, *Advanced Quantitative Analysis*, p. 218, D. Van Nostrand Company, Princeton, N. J., 1943.
6. N. H. Furman (Ed.), *Standard Methods of Chemical Analysis*, 6th ed., p. 1162, D. Van Nostrand Company, Inc., Princeton, N. J., 1962.
7. L. Woehler and O. Balz, Determination of the Valence of Iron, Cobalt, Nickel, Copper, Manganese, Tin, and Tungsten from Their Water Vapor Equilibriums and the Dissociation Pressures of Their Oxides, *Z. Electrochem.*, 27: 406-419 (1921).
8. R. A. Chalmers, *Comprehensive Analytical Chemistry*, Vol. 1c, p. 598, C. L. Wilson and D. W. Wilson (Eds.), American Elsevier Publishing Company, Inc., New York, 1962.

Section
II
Isotopes and Radiation Technology

Isotope Technology Development

Industrial Measurement Considerations for Use of Low-Energy Photons from Nuclear Sources

By Howard J. Evans*

Abstract: *The applications of low-energy photons from radioisotopes for industrial measurement and control are reviewed. The choice of source, the interaction between the photons and the object measured, and the means of radiation detection are considered. Competing alternative techniques are compared. The sum total of effects and optimum choice of equipment are related as they pertain to the desired measurement.*

In this discussion, low-energy photons are defined as those of energies below 200 keV. The photons considered will be either (1) gamma rays or (2) X rays generated by beta rays, photon fluorescence, or some internal mechanism within the atom at the time the X-ray photon is created. The X rays produced by accelerated charged particles impinging on an anode are designated as either machine-generated X rays or X rays from tubes. Industrial applications refer principally to (1) on-line process-control uses where source, sensor, and first stages of the electronics are near the product line and (2) some laboratory uses of low-energy photons. Thus normal factory environmental conditions and space limitations are assumed to prevail at all times for category (1). Likewise, industrial production attitudes are assumed to be dominant in the acceptance and use of the equipment supplied for any measurement. Thus the first consideration will invariably be the effect of the equipment on the producer's profit and loss for the product line. Maintenance costs, downtime, reliability, installation costs—all these factors are very different in on-line application from what they are in a laboratory environment. The liquid-nitrogen-cooled solid-state detector of lithium-drifted germanium found in almost every nuclear laboratory has thus far found little on-line application. The user is not willing to accept the bulk in the installation nor the fact that the detector will be ruined if the low-temperature coolant is lost.

The designer who wishes to use low-energy photons for on-line applications must acquire personnel who understand both chemical and physical principles as well as industrial design. Physicists and/or chemists with a nuclear background, electrical engineers, mechanical engineers, and probably most important of all, field-application engineers must be included. The measurement made will probably be only part of a set of data to be processed by a computer. All considerations must be weighed against alternative approaches. The scientists, therefore, must also be capable of evaluating other physical and chemical interactions useful for the same measurement purposes to avoid development of a good, but noncompetitive, instrument. The low-energy-photon instrument must be superior to other valuable means, in the customer's opinion, for the particular application. He will evaluate the cost of the instrument in terms of increased profits resulting from (1) savings in material, (2) increased throughput, (3) increased quality of product, or (4) a combination of these factors. The instrument for the laboratory is quite different—it need not withstand a hostile environment, and its failure does not stop a production line. It is purchased for use in obtaining increased knowledge for research or development purposes. The laboratory and on-line instrument have one major factor in common—it is not economically feasible to build either one unless there is or will be a sufficient market to make the instrument producer a

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fair profit. An attempt is made in this article to evaluate some of the technical factors that must be considered in developing a low-energy photon measuring system.

Some other techniques or instruments that are being developed or modified for more rapid on-line and laboratory measurements are:

1. Neutron activation analysis.
2. Atomic and molecular absorption.
3. Ion probes [electron microprobe; ion scattering; low-energy electron diffraction (LEED)].
4. Emission spectrograph.
5. Gas chromatography.
6. Ion specific electrodes.
7. Mass spectrometer.
8. Raman spectroscopy.
9. Interaction of beta particles.

Not all these techniques are presently adaptable for on-line use. Automated sampling and processing techniques, even those requiring vacuum, can compete to some extent with on-line processes; hence an awareness of these processes is necessary if a proper judgment is to be made. Comprehensive reviews on items 2, 4, 5, 7, and 8 above, with hundreds of references on each, can be found in the Fundamentals issue of *Analytical Chemistry*.[1] It should be understood that item 2 includes ultraviolet, visible, and infrared radiation. A broad survey on many types of nuclear applications in chemical processes may be found in Ref. 2.

Nuclear-produced low-energy photons offer some unique advantages for on-line process control instrumentation: (1) With the exception of Mössbauer sources, the source output is the result of a nuclear phenomenon that is insensitive to changes in environmental conditions; thus stability is assured. (2) There is no need for an external energy source. (3) The source is small and remains small even when adequately shielded. (4) The source and detector may be hermetically sealed in corrosion-resistant materials, thus making them suitable for use in hostile environments. (5) The interaction of the photons with the material to be measured is completely nondestructive and produces no aftereffects. (6) No contact is required between the measuring system and the material measured. (7) A high degree of reliability is obtainable. (8) The rate of decay is known exactly, so replacement can be either ignored for the long-half-life sources or completely programmed for the shorter-half-life sources.

This paper is neither a review nor a listing of applications of low-energy photons. Many good papers of this type exist; some are cited in the references. The purpose here is to review the type of source–detector–material considerations appropriate for the application of low-energy photons to on-line process-control measurements. There is quite a distinction between techniques suitable for laboratory use and those adaptable to on-line implementation. Much of the latter type of information is proprietary to the individual instrument manufacturer and will not be discussed here.

SOURCES OF LOW-ENERGY PHOTONS

The three primary nuclear sources of low-energy photons are gamma and X-ray emitters, bremsstrahlung sources, and fluorescence sources. Table 1 lists a number of gamma emitters; Table 2, bremsstrahlung sources; and Table 3, Mössbauer radionuclides. It is

Table 1 Low-Energy Gamma Sources

Isotope	Half-life	Energy, keV*	Photons per disintegration†
^{241}Am	433 years	54.9 γ	0.36
		26.4 γ	0.03
		13.9 Np L_α X-ray	0.14
		17.8 Np L_β X-ray	0.18
		20.8 Np L_γ X-ray	0.05
^{109}Cd	1.26 years	87.7 γ	0.039
		22.1 Ag K_α X-ray	0.70
		25.0 Ag K_β X-ray	0.15
^{57}Co	270 days	14.4 γ	0.09
		122.0 γ	0.87
		136.3 γ	0.11
^{155}Eu	1.81 years	105.3 γ	
		86.5 γ	
		60.0 γ	
		45.3 γ	
		26.5 γ	
		18.8 γ	
^{153}Gd	240 days	97.0 γ	
		103.0 γ	
		41.0 Eu K X-ray	
		6.0 Eu L X-ray	
^{125}I	60 days	35.5 γ	0.07
		27.4 Te K_α X-ray	0.88 EC, 0.82 IC
		31.0 Te K_β X-ray	0.20
^{55}Fe	2.7 years	5.9 Mn K X-ray	
^{145}Sm	340 days	61.0 γ	
^{119m}Sn	250 days	24.0 γ	
		25.0 Sn K X-ray	
		4.0 Sn L X-ray	

*X-ray data incomplete.
†Column incomplete.

Table 2 Beta Emitters for Low-Energy Bremsstrahlung Sources

Isotope	Half-life	Maximum energy, keV
^{45}Ca	164 days	254
^{14}C	5.73×10^3 years	155
^{3}H	12.3 years	18
^{63}Ni	92 years	67
^{147}Pm	2.62 years	223
^{151}Sm	~90 years	75.9 (98.4%) 54.3 (1.6%)
^{35}S	88 days	167

somewhat disappointing to find that only about 1% of over 1000 nuclides now potentially available have a significant number of industrial applications. The number of emissions from bremsstrahlung and fluorescence sources will be only a fraction of those from the primary radionuclide because of the low efficiency of the conversion process. For the bremsstrahlung source, an approximate energy of emission, W, from an electron of energy E is given by

$$W = kZE^2 \qquad (1)$$

where $k = 0.7 \times 10^{-4}$, Z is the atomic number of the material decelerating the electrons, and W and E are given in MeV. Since the beta spectrum is usually continuous, the output will be a summation of the continuous photon spectrum for each energy, E, with the low-energy photons strongly attenuated by the source window.

The gamma emitters may be used to produce fairly monochromatic X-ray emissions of lower energy by fluorescence. If the number of disintegrations per gram is high and the internal absorption low, a compact fluorescence source of lower-energy emissions may be produced by placing the primary radionuclide source along the axis of a cavity whose walls are lined with the desired target element. This type and a second source configuration are shown as (*a*) and (*b*) of Fig. 1. There will be some scatter of the primary radiation into the path of the desired fluorescence radiation from both sources. Figure 2 shows the fluorescence yield vs. atomic number for the various elements. This yield is the fraction of electrons that make a fluorescence radiative transfer in filling the vacant K shell.

In industrial applications, encapsulation of the source material becomes of paramount importance,

Table 3 Mössbauer Radionuclides

Z	Mössbauer nuclide	Gamma-ray energy, keV
19	^{40}K	$E = 29.4$
26	^{57}Fe	$E = 14.4$
28	^{61}Ni	$E = 67.4$
32	^{73}Ge	$E = 67.0$
36	^{83}Kr	$E = 9.3$
44	^{99}Ru	$E = 90.0$
50	^{119}Sn	$E = 23.8$
51	^{121}Sb	$E = 37.2$
52	^{125}Te	$E = 35.6$
53	^{127}I	$E = 57.6$
53	^{129}I	$E = 27.72$
55	^{133}Cs	$E = 81.0$
63	^{151}Eu	$E = 21.6$
63	^{153}Eu	$E_1 = 97.4$ $E_2 = 103.2$
64	^{155}Gd	$E_1 = 60.0$ $E_2 = 86.5$ $E_3 = 105.0$
64	^{157}Gd	$E = 64.0$
64	^{158}Gd	$E = 79.5$
65	^{159}Tb	$E = 58.0$
66	^{161}Dy	$E_1 = 25.6$ $E_2 = 74.5$
67	^{165}Ho	$E = 95.0$
68	^{166}Er	$E = 80.6$
69	^{169}Tm	$E = 8.41$
70	^{170}Yb	$E = 84.2$
70	^{171}Yb	$E_1 = 66.7$ $E_2 = 75.9$
70	^{172}Yb	$E = 78.7$
70	^{174}Yb	$E = 76.5$
72	^{176}Hf	$E = 88.3$
72	^{180}Hf	$E = 93.3$
74	^{182}W	$E = 100.1$
74	^{183}W	$E_1 = 46.5$ $E_2 = 99.1$
74	^{184}W	$E = 111.2$
74	^{186}W	$E = 122.5$
76	^{186}Os	$E = 137.2$
77	^{191}Ir	$E_1 = 129.4$ $E_2 = 82.4$
77	^{193}Ir	$E = 73.0$
78	^{195}Pt	$E_1 = 98.9$ $E_2 = 129.7$
79	^{197}Au	$E = 77.3$
91	^{231}Pa	$E = 84.2$
92	^{238}U	$E = 45.0$
93	^{237}Np	$E = 59.5$

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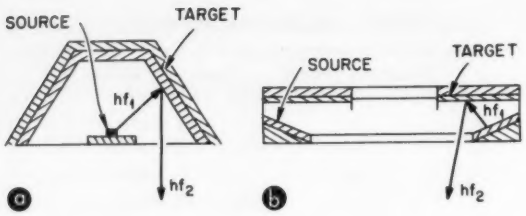

Fig. 1 Source of fluorescence monochromatic radiation. (a) Source and conical target. (b) Flat source and flat target.

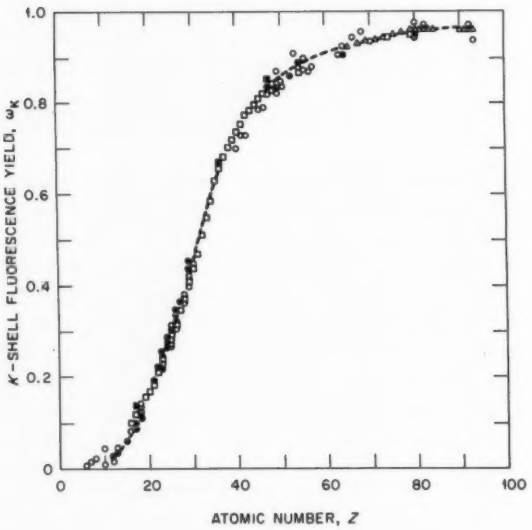

Fig. 2 Fluorescence yield vs. atomic number. □, theoretical, from E. J. Callen, Preprint 1963, unpublished; also, *Bulletin of the American Physical Society*, 7: 416 (1962). △, theoretical, from M. A. Listengarten, *Izvestiya Akademii Nauk Armyanskoi SSR, Seriya Fizika*, 24: 1041 (1960). ■, theoretical, R. A. Rubenstein and J. N. Snyder, *The Physical Review*, 97: 1635 (1955). ○, experimental values. ●, precise experimental values. (After R. W. Fink, by permission.)

especially if the primary source material is an alpha emitter. Since there is always the danger of fire, the source needs to be encapsulated in materials resistant to high temperatures. Proper encapsulation becomes difficult for the lower energies since windows of sufficient integrity may cause excessive attenuation of the emission. One way to circumvent this problem is to use sources of fluorescence radiation described above. Only the primary source, which emits relatively high-energy photons, need be encapsulated. The other window can be left open or sealed by a low-Z material.

PRINCIPLES: INTERACTION OF PHOTONS WITH MATERIALS

Four methods find considerable use in low-energy photon measurements: (1) transmission attenuation, (2) backscatter, (3) fluorescence, and (4) Mössbauer.

Transmission Attenuation

The attenuation of a collimated narrow beam through a thin material of thickness x (Fig. 3*a*) is given by

$$N = N_0 e^{-\mu\rho x} \tag{2}$$

where N_0 represents the intensity of the incident beam; N, the intensity of the exit beam; ρ, the density of the material in g/cm^3; μ, the attenuation cross section in cm^2/g; and x, the thickness of the layer in cm. Values for μ have been tabulated[3] for all the elements. Equation 2 is valid for a narrow beam where, as a result of the interaction between the photon and the material, the photon is either absorbed or scattered out of the narrow beam. The absorption will be by the photoelectric effect whereas the scattering will be chiefly by Compton inelastic scattering and elastic coherent scattering. Values for all three interactions are given in Ref. 3. That value of x which makes $\mu\rho x$ equal to 1 is known as a *characteristic length*. It is custom-

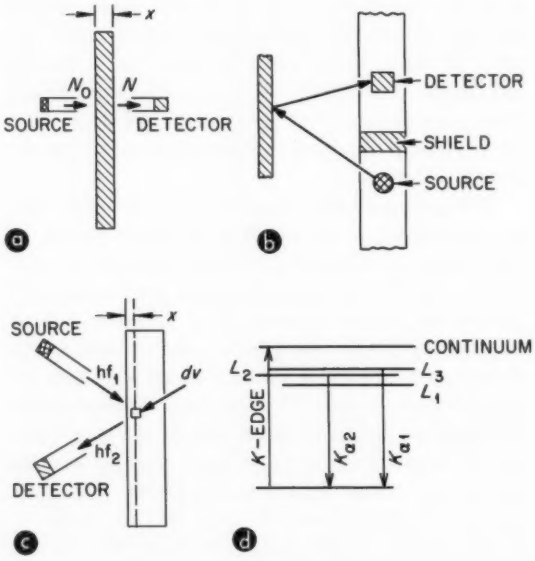

Fig. 3 Uses of low-energy photons. (a) Narrow beam attenuation. (b) Backscatter detection. (c) Fluorescence. (d) Relative energy levels (levels between L and continuum not shown).

ary, when possible, to choose a source of energy that will provide two or three characteristic lengths in the thickness of the material to be tested.

The photoelectric atomic cross section τ is a function of the atomic number of the absorber and of the energy of the incident photon. Theoretically[4]

$$\tau \sim \frac{Z^5}{(E)^{3.5}} \tag{3}$$

Experimentally it is found that the relationship above the K edge of the material is more like the fourth power for the atomic number and the third power for the energy. In the heavier elements, 80% of the photoelectric absorption is by the K shell. The photoelectric cross section begins to dominate below 50 keV for aluminum and below 500 keV for lead. Thus it is always possible to secure relatively thin collimators and shields for energies below 200 keV, as is shown by the value of the $\mu\rho$ product for uranium which is 25 cm^{-1} for a 200-keV photon.

Photons may interact with an electron of an atom in an inelastic scattering phenomenon known as *Compton scattering*. The difference in wavelength in angstroms between the incident photon of wavelength λ_0 and the scattered photon of wavelength λ is given by

$$\Delta\lambda = \lambda - \lambda_0 = 0.0236(1 - \cos\phi)\text{Å} \tag{4}$$

where ϕ is the angle between the direction of the incident and scattered photon. This difference is independent of the energy of the incident photon and depends only on the angle of scattering and can never exceed 0.0472 Å. Thus Compton scattering causes an insignificant change in energy at long wavelengths. The energy T of the recoiling electron is given by

$$T = h\gamma_0\left[\frac{(1 - \cos\phi)\alpha}{1 + (1 - \cos\phi)\alpha}\right] \tag{5}$$

where $\alpha = \dfrac{h\gamma_0}{m_0c^2}$

m_0c^2 = 511 keV

h = Planck's constant

γ_0 = frequency of the incident photon

There also may be coherent scattering—what is commonly called diffraction. This cross section becomes large at low temperatures for a single crystal and wavelengths comparable to or longer than the distance between crystal atomic planes. In this phenomenon the individual photon interacts with the atomic array with negligible momentum transfer and thus produces elastic scattering.

Backscatter

Multiple scattering or individual scattering events in which ϕ is larger than 90° will produce photons that are backscattered from the material (Fig. 3*b*). The formulas for backscattering are quite complex.[5] For low-Z materials and energies in which the Compton effect dominates, the backscattering increases with Z until Z reaches a value at which the photoelectric effect dominates. Further increase in Z then decreases the backscatter due to the fourth-power absorption by the photoelectric effect.

Fluorescence

Fluorescence radiation is depicted in Fig. 3*c* as photons of energy hf_2. As the frequency f is raised, the energy level reaches that of the K edge where the energy of the photon is high enough to remove an electron from the K level and raise it above the Fermi level of the material. Since there is a large number of vacant states above the Fermi level, there will be a sudden increase in photoelectric absorption. The absorption of the photon and the ejection of a photoelectron take place in 10^{-13} sec or less. Within an elapsed time[6] of 10^{-12} to 10^{-16} sec, an electron falls from one of the L shells and replenishes the K shell. Radiative transitions from shell L_{III} to K produce K_{α_1} radiation and transitions from L_{II} to K produce K_{α_2} radiation. This fluorescence radiation is characteristic of the irradiated element and produces the line spectra in X-ray radiation. The percentage of transitions which are radiative is given in a curve by Fink et al.[7] A more recent curve supplied by Fink (to be published) is shown in Fig. 2. The fluorescence yield from an elemental volume dv of Fig. 3*c* is the product of (1) the attenuation of the incident beam in passing through a distance x, (2) the density and photoelectric absorption coefficient for the material and photon energy hf_1, (3) 0.8ϵ, where ϵ is the fluorescence yield obtained from Fig. 2 and 0.8 is the fraction of radiation interacting with the K shell, and (4) the attenuation of the fluorescence radiation in passing through the thickness x. Since both beams are divergent, the two solid angles for the incident and emitted radiation, respectively, are computed. The total emitted radiation is obtained by integrating, over x, the thickness from zero to a depth where a further increase produces a negligible increase in output. This thickness

XUM

is referred to as "infinite thickness." These computations and a yield curve for zinc fluorescence radiation for a layer of zinc on steel have been given by Shiraiwa and Fujino.[8] They carried out their computations over very large solid angles. In instrumentation applications one normally restricts the solid angle by collimation. This decreases the contribution from the slant rays and thus gives a greater depth for an "infinite thickness" than that shown in their curve. Complete data on the K-edge energies and on the various fluorescence emissions may be found in Refs. 3, 9, and 10.

Mössbauer Effects

Some radioactive atoms that are imbedded in a crystal emit energy with no apparent recoil. The emission from these recoilless atoms has a near normal line width. The line width of the spectrum is independent of the temperature of the crystal, but the number of atoms experiencing recoilless transitions is an inverse function of the temperature. The probability, p, for recoilless emission contains the same factor as that for elastic scattering in the Bragg diffraction, namely,

$$p \propto e^{-\frac{\mathbf{K}^2}{3}\langle D^2\rangle} \tag{6}$$

where $\mathbf{K}$ is the wave vector of the photon and $\langle D^2\rangle$ is the mean-square displacement of the atom including both thermal and zero-point motion. This effect is known as the Mössbauer effect and is closely related to Bragg diffraction.

The recoilless fraction, f, in the Mössbauer effect is strongly temperature dependent. The fraction f for a Debye model of the phonon spectrum for a solid of cubic symmetry is given[11] by

$$f = \exp\left\{\left[-\frac{3E_R}{2k\theta_D}\right] \times \left[1 + 4\left(\frac{T}{\theta_D}\right)^2 \int_0^{\frac{\theta_D}{T}} \frac{x}{e^x - 1}\,dx\right]\right\} \tag{7}$$

Here $E_R = E_\gamma^2/2mc^2$, where E_γ is the energy of transition, m is the mass of nucleus, and θ_D is the Debye temperature.

For $T/\theta_D \to 0$,

$$f \approx \exp\left\{\left[-\frac{E_R}{k\theta_D}\right]\left[\frac{3}{2} + \left(\frac{\pi T}{\theta_D}\right)^2\right]\right\} \tag{8}$$

For $T \ll \theta_D$,

$$f \approx \exp\left[-\frac{3E_R}{2k\theta_D}\right]$$

For $T \geqslant \theta_D/2$,

$$f \approx \exp\left\{-\frac{6E_R T}{k\theta_D^2}\right\}$$

When the Mössbauer recoilless atom is an atom in a host crystal of different atomic weight, an effective Debye temperature, Θ_D', is obtained by

$$\Theta_D' = \Theta_D\left(\frac{M_h}{M_m}\right)^{1/2} \tag{9}$$

where M_h is the mass of host atom and M_m is the mass of Mössbauer atom. The Mössbauer effect provides the smallest line width now available for instrumentation—10^{-8} eV or a fractional line width on the order of 10^{-13}. This is so sharp that small Doppler effects are readily measurable. In electrical terminology, one may think of a resonant cavity whose Q is the order of 10^{13}. This exceeding selectivity can be used to measure the effects of the chemical binding of the Mössbauer atoms in a compound since changes in the valence states produce a small change in the nuclear states. The energy shift is measured by producing a Doppler effect and computing the energy shift from the velocity difference between the source and the absorber. The most commonly used source is ^{57}Co with ^{57}Fe as the absorber. Most other Mössbauer materials except tin and iodine must be cooled in order to obtain sufficient emission. Iron-57 is also used because the Mössbauer absorption cross section is about 200 times the photoelectric cross section for the same energy. The energy, however, is only 14.4 keV, so it is quite readily absorbed by the photoelectric effect; applications are therefore restricted to surface or thin-film measurements.

DETECTORS

Energy Resolution

A number of techniques are used to obtain energy resolution of the photons received by a detector. The Mössbauer effect is by far the most sensitive since it can resolve 10^{-8} eV. This is followed by the crystal spectrometer which can resolve a full width at half maximum (FWHM) of 0.1 eV at the long wavelengths.

Line centers can be determined within 0.001 of a FWHM. Angles have been measured to 0.01 sec, although the normal error[10] in reading a dividing circle is 0.1 sec. Today the recommended primary standard wavelength is the $K\alpha_1$ radiation from tungsten which is given as $WK\alpha_1$ and equals 0.2090100 ± 5 ppM Å measured by the best spectrometers. Next in resolving capability at low energies (1000 eV) are the gas proportional counter and the semiconductor solid-state detector, with the gas proportional counter holding the advantage at room temperature and the solid-state detector at low temperature. The proportional counter is used at very low energies. Both the proportional counter and the solid-state detector must use, as an adjunct, an electronic pulse-height analyzer to determine the photon energy.

A band-pass energy-filtering technique normally referred to as Ross filtering[12] utilizes one or two detectors and filters. If two detectors are used, one is covered with a material whose K-absorption edge is just above the photon line spectrum and the other with a material whose K-absorption edge is just below the photon line. The difference signal then is proportional to the intensity of the photon line. If only one detector is used, the two filters are used alternately and for equal counting times. The difference count is again proportional to the line intensity. Ross filters will experience interference from the elements whose L or M radiation falls between the two absorption edges of the filters.

A fifth technique utilizes a crystal or plastic fluorescent disk in conjunction with a multiplier phototube (MPT). This combination has poor energy resolution at low energies but can cover the whole energy range and has the advantage of being fast.

Statistics and Precision

The emission of photons, their interaction, and their detection involve statistical processes. An excellent discussion on this subject is given by Joyce in Ref. 13. In many cases the distribution can be represented with sufficient accuracy by a random Poisson distribution. In this case the standard deviation, σ (square root of the variance), of the total count, N, per unit of counting time is $\sqrt{N}$. This is frequently referred to as "1 σ" and means that a measure of the counts will fall within $(N \pm \sqrt{N})$ sixty-eight percent of the time and that they will fall within $(N \pm 2\sqrt{N})$ ninety-five percent of the time. (The latter is referred to as "2-σ" limits.) Practical operation requires 3-σ limits.

If we take the differential of Eq. 2 and then divide both members by N, we obtain

$$\frac{dN}{N} = -\mu\rho x \frac{dx}{x} \tag{10}$$

If dN is set equal to $n\sqrt{N}$ where n is the number of σ's, then Eq. 10 becomes

$$N_{min} = \left(\frac{n}{\mu\rho x}\right)^2 \left(\frac{x}{dx}\right)^2 \tag{11}$$

In this case N_{min} is the minimum number of counts needed for the precision required, dx/x.

The equation for backscatter fluorescence yield, N, will have the form

$$N = C\left(1 - e^{-bx}\right) \tag{12}$$

where C and b are constants that depend on the solid angles, the conversion efficiency, and the attenuation coefficients for the photons involved. As saturation is approached, $bx \gg 1$, and we have

$$N_{min} \simeq \left(\frac{n}{bx}\right)^2 e^{2bx} \left(\frac{x}{dx}\right)^2 \tag{13}$$

Equations 11 and 13 can be used to determine the counting rate required for precision of measurement desired. Of course there are other factors in the system of a mechanical and electronic nature which degrade the actual performance. Good design is effected by lowering the largest terms in the variance.

There is no universal "best" detector—the optimum choice depends upon the conditions imposed by the application. As indicated in Eqs. 10 and 12, the sensitivity (the ratio of the change in output response to the change in process variable) and the desired precision of measurement set a minimum number, N_{min}, of photons that must be detected per unit interval of time. This requirement on rate alone often forces one to a system that does not detect individual photons. Another important factor is the ambient temperature, although it is frequently possible to use a temperature sensor and then produce the desired temperature at the detector. A third dominant factor is the energy of the photon to be detected. Books[14,15] on nuclear radiation detectors have complete chapters on each of the various types. The material presented here will not be on the theory of the individual detector operation but on some of the limits of operation.

XUM

Ionization Chambers

Where large values of N_{min} per unit time are required, the ionization chamber still finds numerous applications. It is quite rugged mechanically and stable with respect to ambient temperatures. A number of gases may be used to provide a range of sensitivities for various photon energies. It is a volume detector—a characteristic that can be used to advantage in providing an output signal that is insensitive to small displacements of the material being measured (flutter). Large areas of interaction or large-solid-angle detection pose no problem. This is a decided advantage when the measurement supplies a limited number of quanta per unit solid angle. High stability, large dynamic range, long life, and low cost of construction are also favorable attributes. The long life and stability originate in the homogeneous nature of the interacting medium and in its self-restoring properties. As normally constructed it has a time constant of 1 msec or longer, and it does not supply directly any energy analyses of individual photons. For long-life, trouble-free service where only the net value of the signal from large quanta rates is of interest, the ionization chamber will probably continue to be the instrument of choice.

Proportional Counters

The proportional counter is frequently used, especially at low energies, to measure the energy of individual photons or in cases when the counting rate needed is higher than that provided by Geiger–Mueller tubes. The proportional counters usually consist of a cylindrical cathode around a small, coaxial, central anode. Typical values might be a cathode diameter of 0.8 in. and an anode diameter of 0.001 in. Frequently "P-10" (90% argon + 10% methane) is used as the gas. Ion multiplication usually takes place within a short distance of the anode (~0.004 in.), thus making the response uniform over the volume of the detector. The gain is a function of the field strength and gas pressure. Variations of 1% in the center electrode radius can make a variation of 8% in the gain. Other sources of fluctuation in the signal are gas impurities and amplifier noise.

The mean pulse, $\overline{p}$, supplied by a proportional counter is given by

$$\overline{p} = \overline{N} \cdot \overline{A} \tag{14}$$

where $\overline{N}$ is the mean number of ion pairs per incident photon event and $\overline{A}$ is the mean amplification factor per electron. If the ionization and the amplification are independent,[16] then

$$\left(\frac{\sigma_p}{\overline{p}}\right)^2 = \left(\frac{\sigma_N}{\overline{N}}\right)^2 + \frac{1}{\overline{N}}\left(\frac{\sigma_A}{\overline{A}}\right)^2 \tag{15}$$

where σ_p, σ_N, and σ_A are the standard deviations. Let $P(A)dA$ be proportional to the number of single electron amplification events whose amplification factor A lies between A and $(A + dA)$. If one assumes that the ionizing coefficient decreases as a function of the avalanche magnitude in a cylindrical configuration, one arrives at the Polya distribution of form

$$P(A) = A^{[(1/b)-1]}\, e^{-A} \tag{16}$$

with a relative variance of

$$\left(\frac{\sigma_A}{\overline{A}}\right)^2 = b + \frac{1}{\overline{A}} \simeq b \tag{17}$$

where b is usually between 1 and 0.5. The intrinsic ionizing process is related to the Fano factor, F, by

$$\left(\frac{\sigma_N}{\overline{N}}\right)^2 = \frac{F}{\overline{N}} \tag{18}$$

where

$$F = \left(\frac{\sigma_N}{\text{Poisson } \sigma}\right)^2 \tag{19}$$

by definition. The value of $\overline{N}$ can be determined from

$$\overline{N} = \frac{E}{q} \tag{20}$$

where E is the energy of the incident photon and q is the energy required to form an ion pair in electron volts. Introducing Eqs. 17, 18, and 20 into 15, one obtains

$$\frac{\sigma_p}{\overline{p}} = kE^{-\frac{1}{2}} \tag{21}$$

There is some disagreement in the literature over the validity of the Polya distribution, but Eq. 21 is approximately true in most cases. For P-10 gas the value of k usually will lie between 5.7 and 4.7, and F and q will have the values of 0.2 and 26.5 eV, respectively.

If only counting is desired, then the resolving time can be made less than 1 μsec. This detection is based on the rise time of the pulse from collection of the electrons. If accurate energy measurements are to be made, then one must also collect the positive charge. This increases the resolving time by a factor of ~10. Stable operation for high counting rates for an appreciable period of time requires continuous flow-type equipment.

Solid-State Semiconductors

Solid-state semiconductors find wide use in nuclear laboratories and limited use in on-line photon detection applications. In many industrial applications it is necessary to have appreciable distance between the source and the detector. The sensitive area of the semiconductor detector coupled with this distance consideration creates a small solid angle for detection purposes which is often a decided disadvantage. The other disadvantage, at present, is the low band gap and atomic number of the semiconductor material—the first factor requires cooling, and the second limits efficient detection to low photon energies. As one lowers the photon energy, the factor $\mu\rho x$ for a 2-mm value for x for silicon does not attain the value of 1 until the photon energy is decreased below ~25 keV. Thus a silicon detector is inefficient for the higher energies.

The advantages of a semiconductor detector over a gas detector for photons are 5 to 10 times as many unit charges per ionizing event and a denser interacting medium. The rise time can be from microseconds to tens of nanoseconds. Two types of detectors find various applications: the surface barrier and the diffused junction. The conventional type of silicon surface barrier detector has a gold layer (about 40 $\mu g/cm^2$) on a very thin p-type layer on the input side of an n-type high-purity silicon wafer. The gold serves as the negative electrode, and the positive contact is made through a nonrectifying metal contact to the n-type silicon backing. For an abrupt junction the depletion depth D in micrometers is given by

$$D = 0.5\left(\rho_n V\right)^{1/2} \tag{22}$$

where ρ_n is the resistivity of the n-type layer in ohm-cm and V is the applied inverse voltage in volts. The electric field is a maximum at the input surface and decreases linearly to zero with distance to the extent of the depletion layer D. The maximum electric field at the surface in V/cm is

$$E_{max} = 4.2 \times 10^4 \left(\frac{V}{\rho_n}\right)^{1/2} \tag{23}$$

and the capacitance in picofarads for a diode area of A cm^2 is

$$C = \frac{21 \times 10^3 A}{(\rho_n V)^{1/2}} \tag{24}$$

The noise of the preamplifier increases with increase in C; hence it is desirable to make D large. In conflict with this relation is the fact that the internally generated noise in a detector increases with the volume of the detector.

The lithium-drifted detectors have provided a partial answer to the absorption problem by providing a means of obtaining thick junctions. Many articles and reviews exist on this subject, but the use of lithium-drifted germanium detectors is omitted here since they require cryogenics.[17-19] Reference 17 reviews Pell's theory and gives experimental procedures and results, while Ref. 19 gives a curve of the relative detection efficiency of a Ge(Li) detector (4-cm^2 area by 5 mm thick) over the photon energy range from 20 to 700 keV.

Some pertinent parameters of silicon are:

- Energy gap = 1.1 eV at room temperature.
- q = 3.6 eV for production of hole–electron pair.
- Electron mobility = $(2.1 \times 10^9/T^{2.5})$ $cm^2/(V)(sec)$ = 1350 $cm^2/(V)(sec)$ at room temperature.
- Hole mobility = $(2.3 \times 10^9/T^{2.7})$ $cm^2/(V)(sec)$ = 480 $cm^2/(V)(sec)$ at room temperature.
- Z = atomic number = 14.

A marked improvement could be obtained for the detection of photons if a material with a higher Z and band gap could be found. In response to this need, work[20,21] has been going on for a number of years on CdTe, which has a room-temperature band gap of 1.51 eV and an atomic number of 48 to 52. At room temperature, 2 kT = $\frac{1}{20}$ eV; hence a change from 1.1 eV to 1.51 eV in the band-gap effects a reduction factor of 3000 [exp (0.41/0.05) = exp (8)] in the number of free carriers thermally generated in CdTe as compared to the number generated in silicon. At the same time, Z has gone from 14 to an approximate value of 50. Since the photoelectric absorption is a function of $\approx Z^{4.5}$, there is an increase in the tau value by a factor of 300. Real problems exist at present with trapping and noise, but the future looks promising. At present the FWHM value is 100 keV at 100°C and less than 8 keV at room temperature. There will be many

XUM

applications for the CdTe detector if it can be adequately developed.

Another semiconductor detector that is under development is the avalanche multiplying diode. Hauser[22] and Keil[23] have considered the threshold energy needed for avalanche multiplication in germanium and silicon and compared it to the value of 1.5 times the band gap frequently assumed for direct band-gap material. Hauser shows that, for indirect band-gap materials such as germanium and silicon, the value can be as low as 1.1 eV. Carrier multiplication begins at two-thirds of the breakdown voltage with a region of finite multiplication similar to the proportional region of a gas detector followed by a steep rise shortly before breakdown. Huth[15,24] has supplied a number of these silicon detectors to industry. He has reported that he can detect 1.5-keV photons at a temperature of 100°C. He estimates that these events inject 400 electrons into the avalanche region. The gain is sufficient to provide detection in a 200-μA leakage current. One advantage of this detector is its short resolving time of less than 10 nsec.

All semiconductor detectors have fundamental limits to the line width of the displayed spectral line. The intrinsic FWHM in eV, ΔE, is[25]

$$\Delta E_{\mathrm{FWHM}} = (F)^{1/2}\ [8(\ln 2)qE]^{1/2} = 2.36(FqE)^{1/2} \qquad (25)$$

where F is the Fano factor, q is the energy of formation of a hole–electron pair, and E is the energy of the incident photon line. The quantity in brackets assumes that the line distribution is Gaussian. For germanium, q and F are 2.98 and <0.08 eV, respectively, while for silicon they are 3.9 and 0.08 eV. Both must be cooled to liquid-nitrogen temperatures if good resolution is to be obtained. Normal leakage current at room temperatures is 0.1 to 1 μA. Leakage current must be less than a nanoampere for good resolution.

Scintillation-Disk Multiplier Phototubes

A counting detector that has found wide application is the scintillation-disk multiplier-phototube (MPT) assembly. The scintillator may be (1) inorganic crystals such as NaI(Tl) or CsI(Tl), (2) organic crystals such as anthracene or stilbene, or (3) one of several plastics produced by companies such as Pilot Chemicals, Inc., Thorn Electronics, or Nuclear Enterprises. The inorganic crystal NaI(Tl) is widely used because of its relatively large light output and high atomic number; CsI(Tl) has approximately the same atomic number for absorption but produces about one-third of the pulse height. The pulse height–time constant product for the two are about equal, so they produce the same light at room temperature. The CsI(Tl) scintillator is not hygroscopic as is NaI(Tl) and is not brittle but pliable. Anthracene has about one-half of the pulse height and less than 7% of the light output of NaI(Tl), while stilbene and the plastic scintillators have about 1% of the light output of NaI(Tl). The NaI(Tl) crystal has a decay time of $\frac{1}{3}$ μsec, while anthracene and stilbene have decay times of 29 and 5 nsec, respectively. The decay time is the duration of the signal from 70% maximum to 7% maximum. Most of the plastics have two decay components—an exponential type falling to $1/e$ in 2 to 5 nsec and a long-term component. Yates and Crandall[26] exhibit decay curves for 15 crystalline, plastic, and liquid scintillators. The NaI(Tl) scintillator[27] light-output efficiency vs. input photon energy varies as much as 15% between 6 keV and 200 keV, with a maximum efficiency in the range from 10 to 60 keV. There is apparently a color shift below 6 keV since phototubes with different photocathodes show different responses below that energy.

The line-width relative variance V_A at the anode of the MPT is given by[28]

$$V_A = V_t(E) + I(E) + \frac{1 + V_m}{nt} \qquad (26)$$

where $V_t(E)$ = relative variance of light transfer
$I(E)$ = intrinsic relative variance of light production
V_m = relative variance of the MPT
n = mean number of photons produced in the absorption of the photon of energy E
t = probability that a produced light photon will supply a photoelectron at the first dynode of the MPT

If the scintillator is to absorb energies over a large dynamic range, then $V_t(E)$ will be a function of the energy because the high-energy photons will be absorbed throughout the crystal while the low-energy photons will be absorbed in the front layer. In NaI(Tl) there is an anomaly at the iodine K edge. The relative variance V_A increases by a factor of 10 in going from 200 keV to 6 keV with an EMI9536A MPT. The MPT contributes about one-half of the relative variance at an energy of 80 keV, slightly more below, and slightly less above.

Recently there has been a major advance in design.[29-31] By using a wide band-gap, heavily doped p-type semiconductor (GaP) with an electropositive

coating (cesium) as the first dynode, Simon et al.[29] have been able to obtain secondary emission ratios as high as 130 at 2.5 keV. This type of emitter is known as a negative electron-affinity emitter and produces a high secondary-emission ratio because the lifetime of the secondary electrons in the emitter has increased by two orders of magnitude—from 10^{-12} sec to 10^{-10} sec. There is good agreement between the measured yield and that computed from theory. In application the voltage is lowered until the secondary emission ratio is ~30 to 50. This permits an easy separation between one, two, three, etc., electron photoemissions. Since the thermal emissions will be single electron emissions while the photo pulses will be multiple electron emissions, it becomes possible to set a threshold that discriminates against the noise from the thermal emission of the photocathode. (This dynode also permits distinction between one and multiple photon-production processes in nonlinear interactions with laser beams.)

The normalized FWHM of the pulses at the anode for a MPT from a uniform set of light flashes that produce a mean value of N_e photoelectrons at the cathode is[31]

$$\text{FWHM} = 2.36 \frac{1}{\sqrt{N_e}} \left[1 + \frac{kG_2}{G_1(G_2 - 1)} \right]^{1/2} \quad (27)$$

where G_1 = gain of first dynode
G_2 = gain of other dynodes
2.36 = conversion factor between FWHM and standard deviation for a Gaussian distribution
k = experimentally determined constant having a value of about 1.6

With the new dynode the factor within the brackets becomes approximately 1, which means that the multiplication process (gain) has approximately zero variance for this MPT. There remains the variance in the conversion process from photons to photoelectrons at the photocathode.

APPLICATIONS

There are hundreds of applications covering transmission attenuation, backscatter, fluorescence, Mössbauer, tracers, etc. They range from measuring thicknesses of less than milli-inches in film coating on computer storage disks to feet in hydrocarbon fuel containers. In general, beta particles are used for multiple, varying composition because they have low sensitivity to a change in Z, while gamma rays exhibit a high sensitivity to a change in Z. If the composition is relatively stable, then gamma rays may readily be used to measure the ρx product or either factor if the other is held constant. Gamma-ray sources are not only stable but are exceedingly compact because no external power is required and shielding is very simple below 200 keV. Machine-generated photons of 200 keV create an awkward source-space problem due to the high-voltage insulation requirements. To obtain adequate output, the anode-to-cathode voltage (in kV) needs to be approximately 50% higher than the desired photon energy. There is also need for a grid-control output to avoid output control by modulation of the high voltage. Variation of the filament temperature in temperature-limited emission is too slow for adequate high-speed control of the output. Another factor strongly in favor of radioisotope sources is the stable emission rate—even in a high-temperature, corrosive environment. This factor cannot be emphasized too strongly in on-line industrial applications.

The lower limit of industrial on-line applications is about 5 keV because of the absorption problem in materials and in the source–detector apparatus. Laboratory equipment for more basic measurements begins at about 25 keV and extends downward a thousandfold. Only Mössbauer equipment and X-ray spectrometers have adequate energy resolution for many basic studies, with the Mössbauer effect having an order of magnitude better resolution on valence-band effects. Although Table 3 lists a large number of Mössbauer isotopes, only Fe, Sn, I, Au, and a few rare earths have found much application; ^{57}Fe is used more frequently than any of the others. High-energy resolution obtained by means other than through the Mössbauer effect requires high collimation as is done in precision X-ray spectrometers. This decrease in the solid angle reduces the intensity by four or five orders of magnitude, thereby eliminating isotopic sources as a supplier of photons.

Crystal spectrometers are classified according to the configuration of the crystal: (1) flat with Soller slits, (2) cylindrical for line focus sources, and (3) toroidal for point focus sources. Some crystals and their crystal-plane spacings used for long-wavelength measurements are: ADP(200), 3.75 Å; mica, 9.95 Å; lead laurate (PbC_{12}), 35 Å; and lead melissate (PbC_{31}), 82.54 Å. Many instruments are now automated to make many measurements in succession.[32] It is well known that spectrometers are used to measure lattice spacing to an accuracy of a few parts per

XUM

million, but they may also be used to determine degrees of oxidation.[33] For instance, the K_β line shapes of silicon and SiO_2 are appreciably different. The line shape of SiO_2 is quite symmetrical, indicating that all levels of the valence band are filled and that it is an insulator. The silicon line indicates that it is a conductor as shown by the asymmetry of the line with the short-wavelength side being sharp and the long-wavelength side exhibiting a trailing edge.

In the last few years there has been an accelerated interest in the on-line applications of fluorescence of the K and L shells of elements to identify and make quantitative measurements, especially of thin coatings. Suitable line spectra can be obtained either from a gamma source or by a conversion process by inducing fluorescence with a suitable target.[33] Suitable sources and ranges of target fluorescence energies are (1) ^{241}Am (20 to 51 keV); (2) ^{109}Cd(8.6 to 19 keV); (3) ^{153}Gd (19 to 77 keV); (4) ^{155}Eu (31 to 77 keV); and (5) ^{145}Sm (14 to 32 keV). In many ways, for industrial uses, fluorescence is superior to optical spectral-emission measurements. For instance, fluorescence of the K-band is nondestructive, can be averaged over the desired area, and provides only one line whereas optical ranges provide dozens of lines. This technique can be used down to elements of $Z = 11$ (Na). It finds its primary application in metals but can be used on ores, glass, clays, rubber, and some industrial chemicals. Reference 33, Vols. I and II, and subsequent reports on the same symposium give many applications.

Another interesting application of the fluorescence conversion source is that by Kato.[34] The absorption coefficients for hydrogen and carbon are the same at 21.6 keV. Thus, by using a cell of fixed dimensions and compensating for temperature, the sulfur content of a hydrocarbon such as oil may be measured. With this technique, sulfur content up to 4% can be determined with an accuracy of 0.05%. An appropriate source is obtained by fluorescing the silver K line, which is 22.2 keV.

Counting techniques are used extensively in research and in instrumentation. For on-line applications they still have many limitations at low energies when energy resolution is desired. The semiconductor solid-state junction detectors are small in area and need to be cooled for use with the lower energies. The scintillators have a very low conversion efficiency, a cesium 660-keV photon absorbed in a NaI(Tl) crystal producing only 4000 photoelectrons in the conventional MPT.[31] This is only 6 photoelectrons per keV, and the other scintillators have lower efficiencies. A flow-type proportional counter is required if large counting rates are to be used over long intervals. The need for short response times in the process and high accuracy frequently push the required counting rate above 10^6 events/sec. If the source is monoenergetic and of sufficiently high energy and counting is the only interest, then there is no problem at rates of 10^6 events/sec; a plastic scintillator and fast counting circuits can be used. [It is normally assumed that NaI(Tl) will not be used for rates above 10^5 counts/sec.] The high-energy pulse will be far above the background noise; hence a spread in line width is of no consequence in setting the low-level energy discriminator. If a bremsstrahlung source is used and one wishes only to count, then at high counting rates there will be a problem with restoring the base line for the low-level discriminator. The average output counting rate of N for a system with an average input counting rate of N_0 is

$$N = N_0(1 - N_0 r) \tag{28}$$

where r is the resolving time of the system. In MPT's there is spreading of the pulse due to the variation in transit time of the electrons between dynodes. This will not be significant for pulses of greater than 5 nsec. If we allow another 5 nsec for the electronics and set the resolution at 10^{-8} sec, then at a counting rate of 10^7/sec one is losing 10% of the counts. The need for high speed and accuracy frequently forces one into an integrating mode with some attendant leakage problems, choppers, and/or d-c amplification. Above 10^6 events/sec is a region of degrading performance for counting techniques, especially for continuous spectrum sources. Integrating modes can operate at much higher count rates before saturation sets in. The response curve will not be the same for the two methods of measurement since the integrating technique sums the product of *number of counts per unit energy level* and the *energy per level.* At rates of less than 10^5 events/sec, counting techniques provide an easy means of circumventing the low direct current provided by the integrating method.

Details on actual applications may be found in Ref. 35, and the various symposiums are listed there.

FUTURE

It is always somewhat dangerous to prognosticate. However, the prediction of certain trends can be made with a relatively high degree of assurance. Although the computer is widely used today, its application to

overall process measurement and control is still in its initial stage. Conversely the day of the single independent measurement is probably past. Use of low- and high-energy photons, electron interactions, neutron activation, and many other techniques, instruments, and methods will be selectively combined to provide a better and more economic product. This was impossible before the days of the computer because many of the variables are not independent, and the matrix calculations in unfolding spectral data were too complicated and too time consuming.

Another factor that has changed is the miniaturization of solid-state functional blocks capable of handling very high frequencies. With these capabilities it becomes possible to measure and compensate for an instability either electronically or through some phase of the computer computation and still keep the equipment size and cost at acceptable levels. These two factors, for which designers have yearned, now place a much greater burden on the judgment factor in selecting the measuring process. To meet this new responsibility, design engineers and scientists must have adequate knowledge of the basic physics, chemistry, and metallurgy of materials and processes. Never before have the established measurement and process-control methods been in such danger of being preempted by a totally different approach in making a measurement. For instance, in optical measurements, further development of a high-speed electronically tunable laser of high spectral purity would probably completely revolutionize optical spectrometers and chemical and physical material measurements made with light.[36] In transmission or backscatter spectrums of products, deviations of *stored output* vs. *frequency* curves from those for standard samples could be quickly measured and processed in a computer. The power density per unit wavelength is so high that Raman spectroscopy[37,38] is readily accomplished, thus opening up material measurements not attainable by ordinary optical methods. Raman spectroscopy is quite suitable for measurement in polymer chemistry and biochemistry, fields of the future. The most powerful pulsed laser in the world today has 50 J output in 10 psec. This is 5 TW, which is 500 times the steady-state electrical generating capacity of the United States.[39]

There will continue to be a large number of transmission, backscatter, and fluorescence applications where accuracy of measurement is the dominant factor and rate of measurement is less important. In these applications, isotopic sources of gamma rays and X rays will be used. There are, however, increasing numbers of high-speed operations which, for successful operation, require photon rates equivalent to that from hundreds or thousands of curies. These applications will use the X-ray tube as a source of photons. In continuous operation, the X-ray tube can supply 10^4 to 10^5 as many photons per unit area of emitting surface as the most active isotope source. Pulsed sources can supply instantaneous amounts many times higher, which reach their peak in the field emitters. Since conventional X-ray-tube sources are quite unstable emitters when compared with isotopes, they will be used in (1) applications in which ratio measurements are simultaneously made, thus minimizing the instability factor and (2) applications where a compensatory measurement to correct for the instability can be made. New technology will open up many applications in category (2).

X-ray photography has had wide use as a nondestructive testing technique for many years. The development of semiconductor gamma-ray cameras and the commercialization of low-light-level techniques formerly reserved only for classified programs has opened up new applications.[40,41] The silicon-diode-array camera tube developed by the Bell Telephone Laboratories is far more sensitive than film in the 5- to 50-keV photon region (about 60 times at 10 keV). It has storage times of 1 sec at room temperature and up to 1 min when the target is cooled. Scintillators in conjunction with a low-light-level camera also provide a large increase in sensitivity.

There is no question that many applications for fuel and flow measurements on various types of equipment, especially aircraft, could be found if the programs on the CdTe detector can solve the trapping and noise problems. Most of these problems are associated with materials, and there is need for additional applications so that the total market can support sufficient basic studies on properties of the materials. Some encouragement for CdTe is found in its added application as an electro-optic modulator in infrared equipment.[42]

Many of the new applications will be at low energies and at high counting rates, where good energy resolution is desired. This virtually eliminates the proportional counters, limited because of low counting rates, and the scintillators, because of insensitivity to low photon energies. Thus there is real need for the development of the CdTe diode and the silicon avalanche diode for equipment and on-line applications.

Laboratory instrumentation will probably not use isotopic sources except for simple portable instruments

XUM

and for measurements utilizing the Mössbauer effect. The high-energy resolution required makes it mandatory that the source be intense. There is an array of rather closely related techniques and laboratory equipment which will continue to find greater use but probably not directly on line. When used in on-line production applications, the techniques will involve good vacuums. An example is the ion-implantation technique so widely used now in fabrication of semiconductors.[43,44] The same type of instrument can supply ions for channeling studies which provide information on the position of impurity atoms in a semiconductor lattice.[45] These studies will be combined with X-ray studies in metallurgy.[46] Closely akin are the surface studies on emitters, catalyzers, semiconductors, and chemisorbers.[47] The use of energy analysis of scattered electrons (EASE) with low-energy electron diffraction (LEED) has provided the ability to detect the equivalent of 0.02 monolayers of cesium on a silicon surface. To complete the array, one must include the electron microscope[48,49] which today can resolve distances of about 1 Å. These devices will be automated for use in many types of material studies but will probably remain in the laboratory for some time.

An approach that may see considerable application in the future is the incorporation into the manufacturing process of materials whose sole purpose is to permit the making of some kind of measurement. For instance, a sulfur atom has a low atomic number and thus a low absorption coefficient for energetic photons; however, it can be made to attach itself to mercury with a high degree of specificity, as for use in SO_2 measurements. One measures the mercury and infers the amount of sulfur.

In the future much more use will be made of the detail in spectral line shapes and in the unfolding of multiline spectrums. Most of the interactions between very-low-energy photons and materials involve resonant effects.[50] Resonant effects cover frequencies from 10^6 Hz for nuclear magnetic resonance to 10^{18} Hz for Mössbauer effects. In between lie electron spin resonance, microwave resonance, infrared, visible, and ultraviolet. Qualitative measurements are usually made by determining the frequency of resonance and quantitative measurements by the degree of absorption of the line or by the extent of its displacement or distortion. Visible and ultraviolet photons are usually used to measure bond strength in materials or their derivatives while infrared photons are used to determine functional groups.

(FJM, REG, PSB)

REFERENCES

1. *Anal. Chem.* (1968 Annual Reviews), **40:** (1968).
2. P. S. Baker, Radioisotopes in Chemical Processes, *Chem. Eng.*, **75:** 179-186 (1968).
3. E. Storm and H. I. Israel, Photon Cross Sections for 0.001 to 100 MeV for Elements 1 Through 100, USAEC Report LA-3753, Los Alamos Scientific Laboratory, June 1967.
4. J. McConnell, *Quantum Particle Dynamics*, Chaps. IX and X, Interscience Publishers, Inc., New York, 1960.
5. N. N. Shumilovskii and L. V. Mel'ttser, *Radioactive Isotopes in Instrumentation and Control*, R. F. Kelleher (Transl.), P. J. Blaetus and G. A. Young (Eds.), The Macmillan Company, New York, 1964.
6. L. G. Parratt, Electronic Band Structure of Solids by X-Ray Spectroscopy, *Rev. Mod. Phys.*, **31:** 616-645 (1959).
7. R. W. Fink, R. C. Jopson, Hans Mark, and C. D. Swift, Atomic Fluorescence Yields, *Rev. Mod. Phys.*, **38:** 513-540 (1966).
8. Toshio Shiraiwa and Nobukatsu Fujino, Theoretical Formulas for Film-Thickness Measurement by Means of Fluorescent X-Rays, in *Advances in X-Ray Analysis*, Vol. 12, pp. 446-456, Plenum Press, Inc., New York, 1969.
9. J. A. Bearden, X-Ray Wavelengths, *Rev. Mod. Phys.*, **39:** 78-124 (1967).
10. J. A. Bearden and A. F. Burr, Reevaluation of X-Ray Atomic Energy Levels, *Rev. Mod. Phys.*, **39:** 124-142 (1967).
11. Krishan Mahesh, On the Temperature Dependence of the Mössbauer Effect, *Nucl. Instrum. Methods*, **65:** 349-352 (1968).
12. P. A. Ross, New Methods of Spectroscopy for Faint X-Radiations, *J. Opt. Soc. Amer. and Rev. Sci. Instrum.*, **16:** 433-437 (1928).
13. W. B. Joyce, Origin of Counting Statistics in Simple Nucleonic Instruments, *Amer. J. Phys.*, **37:** 489-494 (1969).
14. W. J. Price, *Nuclear Radiation Detection*, 2nd ed., McGraw-Hill Book Company, Inc., New York, 1964.
15. Conference on Semiconductor Nuclear-Particle Detectors and Circuits, Publication 1593, National Academy of Sciences, Washington, D. C., 1969.
16. M. W. Charles and B. A. Cooke, Proportional Counter Resolution, *Nucl. Instrum. Methods*, **61:** 31-36 (1968).
17. H. H. Sher, Lithium–Ion Drift Mobility in Germanium, *J. Appl. Phys.*, **40:** 2600-2607 (1969).
18. W. A. Hackler and C. Kikuchi, Study of Lithium Mobility in Irradiated Silicon, *Nucl. Sci. Eng.*, **31:** 175-182 (1968).
19. D. P. Donnelly and M. L. Wiedenbeck, The Relative Detection Efficiency Calibration of a Ge(Li) Detector at Low Energies, *Nucl. Instrum. Methods*, **64:** 26-28 (1968).
20. W. Akutagawa and K. Zanio, The Possibilities of Using CdTe as a Gamma Spectrometer, 11th Scintillation and Semiconductor Counter Symposium, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-15(3): 266-274 (1968).
21. K. R. Zanio, Improved CdTe for Gamma Detection Between −100°C and +100°C, *Appl. Phys. Lett.*, **14:** 56-58 (1969).
22. J. R. Hauser, Threshold Energy for Avalanche Multiplica-

tion in Semiconductors, *J. Appl. Phys.*, **37**: 507-509 (1966).

23. G. Keil, Silicon Avalanche Diodes as Detectors for Gamma Radiation, *Nucl. Instrum. Methods*, **65**: 277-284 (1968).
24. R. J. Locker and G. C. Huth, A New Ionizing Radiation Detection Concept Which Employs Semiconductor Avalanche Amplification and the Tunnel Diode Element, *Appl. Phys. Lett.*, **9**: 227-230 (1966).
25. D. W. Aitken, Recent Advances in X-Ray Detection Technology, 11th Scintillation and Semiconductor Counter Symposium, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-15(3): 10-46 (1968).
26. E. C. Yates and D. G. Crandall, Decay Times of Commercial Organic Scintillators, 10th Scintillation and Semiconductor Counter Symposium, *IEEE (Inst. Elec. Electron Eng.), Trans. Nucl. Sci.*, NS-13(3): 153-158 (1966).
27. D. W. Aitken, B. L. Beron, G. Yenicay, and H. R. Zulliger, The Fluorescent Response of NaI(Tl), CsI(Tl), CsI(Na), and CaF(Eu) to X-Rays and Low-Energy Gamma Rays, 13th Nuclear Science Symposium, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-14(1): 468-477 (1967).
28. G. H. Narayan and J. R. Prescott, Line-Widths in NaI(Tl) Scintillation Counters for Low-Energy Gamma Rays, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-13(3): 132-137 (1966).
29. R. E. Simon, A. H. Sommer, J. J. Tietjen, and B. F. Williams, New High-Gain Dynode for Photomultipliers, *Appl. Phys. Lett.*, **13**: 355-356 (1968).
30. R. E. Simon and B. F. Williams, Secondary-Electron Emission, 11th Scintillation and Semiconductor Counter Symposium, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-15(3): 167-170 (1968).
31. G. A. Morton, H. M. Smith, Jr., and H. R. Krall, The Performance of High-Gain First Dynode Photomultipliers, 15th Nuclear Science Symposium, *IEEE (Inst. Elec. Electron. Eng.), Trans. Nucl. Sci.*, NS-16(1): 92-95 (1969).
32. P. G. Cath and J. Ladell, Principles and Design of the Automatic Single Crystal Diffractometer System PAILRED, I. Principles of the Diffractometer, II. The Automatic Control Circuits, *Philips Tech. Rev.*, 29(6): 165-185 (1968).
33. J. S. Watt, Some Recent Developments in Low-Energy X- and Gamma-Ray Sources and Applications in Australia, Proceedings of 2nd Symposium on Low-Energy X- and Gamma Sources and Applications, Vol. 2, pp. 663-695, P. S. Baker and M. Gerrard (Eds.), USAEC Report ORNL-IIC-10, Oak Ridge National Laboratory, 1967.
34. M. Kato, Present Status of Research and Applications of Low-Energy X- and Gamma-Ray Sources in Japan, Proceedings of 2nd Symposium on Low-Energy X- and Gamma Sources and Applications, Vol. 2, pp. 723-745, P. S. Baker and M. Gerrard (Eds.), USAEC Report ORNL IIC-10, Oak Ridge National Laboratory, 1967.
35. K. W. Ostrowski, L. Górski, and J. Niewodniczanski, Some Applications of Low-Energy Gamma and X-Ray Sources in Poland, *Nukleonika*, 13(3): 289-300 (in Polish) (1968); USAEC Report AEC-tr-6931/3, pp. 34-45, 1968.
36. M. W. Dowley, Efficient CW Second Harmonic Generation to 2573 Å, *Appl. Phys. Lett.*, **13**: 395-397 (1968).
37. *Anal. Chem.* (1968 Annual Reviews), **40**: 320R-330R (1968).
38. J. L. Koenig, Laser Raman Spectroscopy, *Res./Develop.*, pp. 18-22 (June 1969).
39. Record Breaking Pulsed Laser, *Opt. Spectra*, p. 40 (March–April 1969).
40. T. M. Buck, H. C. Casey, Jr., J. V. Dalton, and M. Yamin, Influence of Bulk and Surface Properties on Image-Sensing Silicon Diode Arrays, *Bell Syst. Tech. J.*, **47**: 1827-1873 (1968).
41. A. N. Chester, T. C. Loomis, and M. M. Weiss, Diode Array Camera Tubes and X-Ray Imaging, *Bell Syst. Tech. J.*, **48**: 345-381 (1969).
42. J. E. Kiefer, Electro-Optic Characteristics of CdTe at 3.39 and 10.6 μ, *Appl. Phys. Lett.*, **15**: 26-27 (1969).
43. J. W. Mayer, Use of Ion Implantation Techniques to Fabricate Semiconductor Nuclear Particle Detectors, *Nucl. Instrum. Methods*, **63**: 141-151 (1968).
44. A. G. Foyt, J. P. Donnelly, and W. T. Lindley, Efficient Doping of GaAs by Se^+ Ion Implantation, *Appl. Phys. Lett.*, **14**: 372-374 (1969).
45. W. D. Mackintosh and J. A. Davies, Rutherford Scattering and Channeling—A Useful Combination for Chemical Analysis of Surfaces, *Anal. Chem.*, **41**(4): 26A-35A (1969).
46. V. V. Nemoshkalensko and V. V. Gorskii, Investigation of the X-Ray K-Emission Spectrum of Aluminum, *Ukr. Phys. J. (Engl. Transl.)*, **12**: 812-817 (1967).
47. R. E. Weber and A. L. Johnson, Determination of Surface Structures Using LEED and Energy Analysis of Scattered Electrons, *J. Appl. Phys.*, **40**: 314-318 (1969).
48. C. J. Rakels, J. C. Tiemeijer, and K. W. Witteveen, The Philips Electron Microscope EM300, *Philips Tech. Rev.*, **29**: 370-386 (1968).
49. P. R. Thornton, D. V. Sulway, and D. A. Shaw, Scanning Electron Microscopy in Device Diagnostics and Reliability Physics, *IEEE (Inst. Elec. Electron. Eng.), Trans. Electron Devices*, ED-16(4): 360-371 (1969).
50. A. M. Stoneham, Shapes of Inhomogeneously Broadened Resonance Lines in Solids, *Rev. Mod. Phys.*, **41**: 82-108 (1969).

XUM

Development of Some Radioisotope Procedures for Measuring Water Movement in Trees*

By Peyton W. Owston, James L. Smith, and Howard G. Halverson†

Abstract *Water transpired by trees greatly reduces the water available to streams from forested watersheds. An attempt is being made in the Sierra Nevada of California to measure this passage of water through individual trees by radioactive tracers. Initial experiments were conducted with lodgepole pine (*Pinus contorta *Dougl.) and red fir (*Abies magnifica *A. Murr.) during different seasons of the year. Phosphorus-32 was injected into standing trees through small lateral roots. The rate of water movement was determined by two measurements: the path of movement was monitored outside the bark with a portable scintillation detector and the cross-sectional pattern of movement was determined by autoradiographing transverse disks of stems.*

Rates of daytime isotope movement for both species ranged generally from 20 to 70 cm/hr during the summer. Movement in winter averaged 0.3 to 1.3 cm/hr in red fir. The fastest rates of ascent, 1.5 to 2.0 m/hr, were measured in early spring when as much as 12 ft of snow lay on the ground. Most of the movement occurred in the outer sapwood, but some radioactivity was found 6 to 8 cm deep at times of rapid ascent.

Lack of information about the amount of water transpired by forest vegetation has hampered the development of equations for determining how much water could be "saved" through timber harvest or brush removal. Studies of watersheds have shown that the amount saved varies from region to region.[1-3] These variations are not unexpected since soils, climate, terrain, and forests differ so much from one watershed to another.

Instead of studying entire watersheds, a more reasonable approach would be to determine the amount of water used by individual trees and to total these amounts on the basis of groups of trees. Water use from these groups can then be combined to estimate water use from entire watersheds. The aim of this study is to correlate the amount of water used by individual trees to environmental factors that affect such use and, from this correlation, to develop an equation for tree water-use that will have widespread application in the Sierra Nevada of California.

The first step in the program is to develop a technique for measuring water movement in individual trees. Forest trees use large quantities of water, but the precise amount has never been determined. Techniques applicable to small vegetation are impractical for full-sized trees, and methods applied previously to large trees have generally created unnatural conditions that make results suspect.

Modern radioisotope techniques may make measuring this water movement possible. Isotopes can be introduced into trees with little or no damage to the closed xylem system, and the rate of upward movement can be followed from outside the bark with portable detectors. The pattern of movement can be determined accurately by use of autoradiographs taken from transverse sections of the trees, and autoradiography of microscopic sections can be used to determine the actual fiber-cavity space used in water transport.

Once the flow rate and the area through which the water travels are known, the amount of water moving past a given point in a stem can be computed for any time span. The problem becomes roughly one of "water flow through porous media."

This article reports a method of using ^{32}P for tracing water movement in lodgepole pine (*Pinus contorta* Dougl.) and red fir (*Abies magnifica* A. Murr.) in the central Sierra Nevada at various times of the year. The first effort was to determine if ^{32}P or ^{131}I could be used successfully in lodgepole pine. The subsequent experiments were designed to refine methods of isotope injection and detection, to determine seasonal pattern of water use by trees, and to find the path of ascent in the tree stems. A series of injections of ^{32}P were made into trees during a cold winter (February 14), a cold spring (April 4), a warm early summer (June 20), and a warm, dry, late summer (September 6). A method of isotope injection was selected after field tests, and an isotope was chosen for future injections. Methodology has been developed for sampling increment cores from the tagged trees and for preparation of microtome sections for autoradiography.

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XUM

LITERATURE REVIEW

In the literature, the methods described to study water use by trees fall roughly into three categories: (1) estimation of long-term evapotranspiration by measurement of meteorological factors[4,5] or soil-moisture depletion,[6,7] (2) direct measurement of water loss from soil or foliage, and (3) measurement of water flow in stems.

Determination by Meteorological Factors

The techniques in the first category have serious deficiencies. The estimation of evapotranspiration by measurement of meteorological factors ignores several important items affecting plant water-use. Although energy-balance equations can be used to estimate potential evaporation of water from foliage, they cannot be used to estimate the actual amount drawn through the tree and delivered to the foliage. The amount depends upon available moisture in the soil, soil temperature, air temperature, solar radiation, and internal tree stresses. In fact, the process by which trees absorb and transport water is not yet fully understood.

Scientists have attempted to estimate plant water-use by measuring soil-moisture depletion in the area adjacent to the plant.[8] Were it not for moisture migration in the soil, this method would be more feasible. However, as fast as moisture is used by a plant, moisture moving from adjacent areas can replace it.

Determination by Water Loss from Soil or Foliage

Techniques of measurement of water loss from soil in containers or from foliage are useful for small trees or individual branches of larger ones. Small trees placed in lysimeters are weighed periodically, or water use is computed by measuring water intake and drainage.[9,10] Not only is this method impractical for large trees, but it often creates unnatural conditions when the roots become bound in the containers.

Transpiration from small trees has been determined by enclosing them in tents and measuring the water vapor given off.[11] This technique also is impractical for large trees. Transpiration rates may be influenced by the "greenhouse effect" caused by the enclosure of the trees.

Another widely used technique is the rapid weighing of leaves detached from trees.[12,13] The assumption is made that, for a short period of time after detachment, a leaf will continue to transpire at the same rate as it did before detachment. The assumption is questionable owing to the rapid release of tension in the xylem system. Furthermore, to convert measurements from isolated leaves or branches to total transpiration values for large trees is most difficult—if not impossible.

Determination by Water Flow in Stems

The third type of measurement has been approached in a variety of ways and should be workable for large trees. Such techniques do not measure actual transpiration rates from foliage, but they can be used to measure the path, speed, and, hopefully, the amount of water that will ultimately be transpired by the trees. Determining the path of water movement[14-16] by tracing dyes has probably been the most widely used method, but the accuracy of such measurements is questionable because direct stem injections disturb the xylem. Also, the tree has to be cut to find the dye.

The thermoelectric, or heat-pulse, technique[17] has been used in various forms since it was first developed in the 1930's. In this technique a pulse of heat is injected into the stem, and its movement is followed by thermocouples placed a short distance both above and below the heat source. This movement is related to sap velocity through the use of the formula $V = 900/t_0$ where V = sap velocity (cm/hr) and t_0 = time elapsed from heat input to completion of reading.[18] The "900" is a constant which varies with thermocouple spacing. It has been refined in recent years,[18-21] but no one has yet related heat-pulse velocity to actual water movement. The technique can be used to estimate the cross-sectional area in which water movement occurs, but it cannot be used to determine the actual fiber-cavity space used in water transport.

In 1932, Hüber[22] first used radioisotopes to measure water movement in tree stems. Using ^{212}Pb as a tracer, he reported that water moved in conifers at rates between 16 and 30 cm/hr. Most of the subsequent isotope studies, including those of recent years, have involved techniques that disturb the xylem system. The basic injection procedure, which will be referred to as the trough technique, has been to place a watertight trough or cup against the tree trunk, fill the container with isotope solution, and cut or drill a hole into the trunk below the surface of the solution. Although the method is a reliable way to get materials into the tree, it does, however, create unnatural uptake conditions by releasing tension in the xylem and providing the stem with a free-water source.

XUM

Ferrell and Hubert[23] used the trough technique in 1952 to inject ^{32}P and ^{45}Ca into western white pine (*Pinus monticola* Dougl.). The ^{32}P rose at an average rate of 20 cm/hr over a period of several days during summer, three to four times as fast as in fall when transpiration conditions were not so good. Ferrell and Hubert also tried a root-absorption technique for injection of the isotopes, but it was not successful.

Fraser and Mawson[24] used ^{45}Ca and ^{86}Rb to study water movement in eastern white pine (*Pinus strobus* L.) and yellow birch (*Betula alleghaniensis* Britton). They used the trough technique, but in some cases they poured isotope solution into holes drilled in the trunks. The isotopes moved upward in the xylem in narrow bands that spiraled dextrally. They moved into lateral branches only where the base of the branch fell along the path of ascent. The ^{32}P rose initially at 1.8 m/hr in the birch. Fraser and Mawson attributed this initial surge to the lowering of tension in the xylem caused by the injection technique.

The trough technique has been used to inject ^{86}Rb into pin oak (*Quercus palustris* Muenchh.). Beckman et al.[25] obtained movement rates of 3 to 4.5 m/hr. Atrokhin[26] found that soil-injected ^{32}P took 6 days to reach the crowns of 30-year-old oak trees. Treating red pine (*Pinus resinosa* Ait.), Postlethwait and Rogers[27] found that ^{32}P rose in straight, narrow paths and around horizontal saw cuts.

Tritiated water would probably be the ideal tracer for studies in tree-water movement except for the impossibility of detecting the substance from outside the bark as it passes up the stems. Despite this drawback it is useful for certain kinds of studies. Wray and Richardson[28] measured rate of movement with tritium by using many small willow seedlings and destructively sampling several stems at successive time intervals. By analyzing the water extracted from frozen stem sections, they found rates of upward movement to be 120 to 140 cm/hr. Lewis and Burgy[29] and Woods and O'Neal[30] used tritium to detect rooting characteristics of oak trees. Tritium uptake was significant in both these studies, but determinations of rate of movement in the tree stems could not be made.

Several other studies using mineral isotopes are discussed elsewhere in this report. They include the soil and root injections of ^{32}P by Moreland[31] and Farrar,[32] the comparative use of four isotopes by Akhromeiko and Zhuravleva,[33] and a comparison of ^{32}P and the thermoelectric method in poplar and birch.[34]

FEASIBILITY TEST

Materials and Methods

The objectives of the first experiment were to find out (1) if lodgepole pine would absorb a radioisotope solution through small lateral roots and (2) if the ascent of the isotope could be measured from outside the tree. Two trees about 10 in. d.b.h.* and 30 ft tall were tested. They were growing on the grounds of the Central Sierra Snow Laboratory, Soda Springs, Calif., on the west slope of the central Sierra Nevada at 6900 ft elevation.

Both a beta emitter and a gamma emitter with relatively short half-lives were tested. Phosphorus-32, a beta emitter with a 14.3-day half-life, was used as carrier-free $Na_3{}^{32}PO_4$ in 0.2 *M* NaCl; ^{131}I, a gamma emitter with a half-life of 8.1 days, was used as carrier-free $Na^{131}I$ in 0.1 *M* NaOH. The ^{32}P was injected into one tree, and ^{131}I was introduced into the other.

A small lateral root from each tree was isolated by digging down about 4 in. into the soil. The roots were clipped at a point where their diameters were about 2 mm, and the roots, which were still attached to the tree, were inserted, in situ, into a vial containing 10 mCi of isotope in 10 ml of solution. Soil was replaced and covered with rock and lead until only background radiation could be detected above ground.

Movement of the tracers was monitored from outside the bark by passing various nuclear detectors around the boles of the trees 1 day and 5 days after injection of the isotopes. The detectors tested were a Geiger–Mueller (G–M) survey meter, a NaI scintillation detector with scaler, and a portable rate meter with a NaI scintillation detector. Until isotope was detected in the stems, monitorings were made around the entire circumference of the trees and from ground level to 150 cm above ground. The detectors were passed as close to the bark as possible without actually touching the trees. Readings were recorded at periodic height intervals along reference lines which corresponded to the cardinal compass directions.

After an isotope path was detected in a tree stem, monitoring was concentrated in the area of the path. The top height of the path was determined periodically to compute the rate of ascent. Width of the path was also estimated with the detector. Lower branches and needles were monitored on the fifth day after injection.

*Diameter at breast height or 4.5 ft above ground.

The movement of the isotope was followed within the xylem by placing a small, custom-built G–M tube, attached to a linear rate meter, into increment core holes. These measurements were made to confirm the presence of isotope as found by outside-the-bark techniques and to estimate the cross-sectional area of movement by shifting the detector to various depths in the increment core holes.

Determinations of cross-sectional transport were also made by taking 5-min counts of serial increment core segments 1 or 2 cm in length. The segments were placed in well-shielded planchets and counted with a G–M tube in a laboratory. The increment cores had been collected at intervals of time and height from the tree at the four cardinal compass directions. Sap velocity was also measured periodically by a heat-pulse meter.

Results

The general weather pattern for the first week of the experiment was not conducive to rapid transpiration. The first 4 days were clear but calm, and midday temperatures ranged from 9°C to 17°C. Net solar radiation during midday varied from 1.0 to 1.3 langleys* per minute for the first 2 days. The fifth and sixth days of the test were cool and cloudy with high humidity. Intermittent precipitation totaled 0.15 in.

Phosphorus-32 moved up the tree in a relatively straight path approximately 8 cm wide. Readings outside the bark indicated that the ^{32}P had reached a height of 10 cm on the morning after the day of isotope injection (Fig. 1*a*). The radioactivity moved a short distance laterally during the day, but the front remained the same height above ground. By the fourth day the ^{32}P had reached a height of approximately 500 cm—still moving in a narrow path (Fig. 1*b*). Thus the average rate of isotope movement for the 4 days and nights was about 5 cm/hr. Readings of heat-pulse velocity during midday ranged from 2.4 cm/hr to 9.5 cm/hr on the day of injection and the following day.

The ^{32}P was detected in the lower main branches along the path of ascent (Fig. 1*b*), but not all the secondary and tertiary branches gave evidence of radioactivity. Similarly, only a portion of the needle fascicles on a "hot" branch contained the tracer.

Counts of increment cores collected after the first week of the test indicated that most of the radioactivity was centered within the inner bark and outer centimeter of sapwood. In measurement made outside

*1 langley = 1 gram calorie/cm^2.

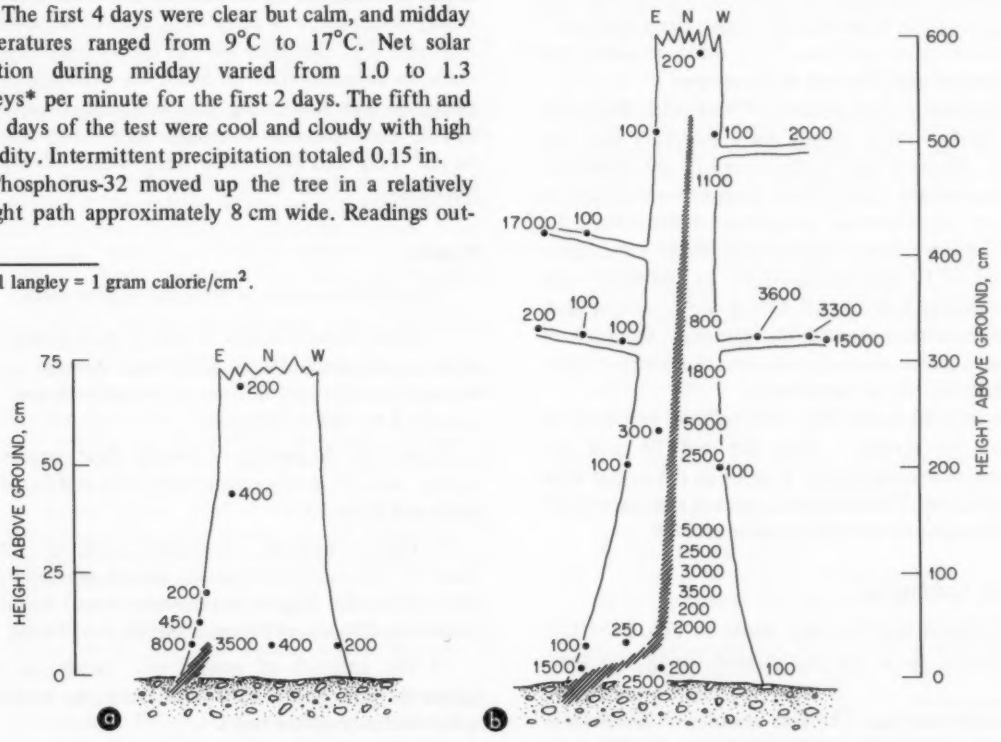

Fig. 1 Schematic diagrams show path of ^{32}P movement in lodgepole pine study tree. Numbers refer to gross counts per minute. Background radiation ranged from 100 to 400 counts/min. Dark shading represents path of radioisotope. Injections made June 2, 1966. (a) Condition on June 3, 1966; (b) condition on June 7, 1966.

XUM

the bark, and from cores, little radioactivity was found in cores taken from positions away from the path of ascent. The activity persisted long after injection date. Cores collected at the end of July—almost 2 months after injection—showed some radioactivity as deep as 5 to 8 cm in the xylem.

The first evidence of radioactivity from ^{131}I in the stem of the second tree was not found until the afternoon of the sixth day of the experiment. Then it occurred only as a small spot extending from 113 to 137 cm above ground. Occasional monitoring during the next several weeks revealed no movement from this position. No radioactivity was found in the lower branches or needles of the tree or in increment cores collected from this tree. The only activity found was in strips of inner bark taken from the small "hot" spot.

FIELD TEST OF TECHNIQUE

Results of the feasibility test showed that ^{32}P would make a reliable tracer for water flow in trees. It was absorbed readily by trees, and its radioactivity was strong enough to be monitored from outside the bark.* A series of field tests was next made to refine the techniques of injection and measurement.

Experiments were conducted four times during the year: winter, early spring, early summer, and late summer. All tests were made on red fir and lodgepole pine trees on the Onion Creek Experimental Forest, a 3000-acre experimental tract about 3 miles from the Central Sierra Snow Laboratory. Each experiment consisted of 10 isotope injections. In addition to the basic techniques developed during the feasibility test, autoradiography was used to determine the path of movement and cross-sectional area of transport more accurately than in the earlier test.

The straight-boled trees selected had fair to good exposure to sunlight. Both the red fir and the lodgepole pine ranged from 8 to 14 in. d.b.h. and were 30 to 60 ft tall. The boles were pruned and marked off in 50-cm segments to 350 cm above ground.

Injection Techniques†

The winter injection was made on Feb. 14, 1967, with 7 ft of snow on the ground, using the same technique that had been used in the feasibility tests. The ^{32}P was introduced on both the north and south sides of two pines and two firs and on the south side of one pine and one fir.

On the basis of earlier tests, 5 mCi rather than 10 mCi of ^{32}P was used. In addition, the chemical compound was changed from carrier-free Na_3PO_4 at *p*H 9.5 to Na_2HPO_4 at *p*H 7.0, with a specific activity of 50 mCi/millimole. Black[35] and others have suggested that plant roots more readily absorb disodium phosphate than trisodium phosphate.

The second experiment was begun on Apr. 4, 1967, when the snowpack was 12 ft deep. The only major change in procedure was that five injections were disodium phosphate at *p*H 7.0, and the other five were trisodium phosphate at *p*H 9.5.

The next injections of $Na_2H^{32}PO_4$ and $Na_3{}^{32}PO_4$ were made on the morning of June 20, 1967. The snowpack was still 1 to 4 ft deep. Five injections of 5 mCi each were made with each chemical.

The fourth and last isotope test of this series was begun on the morning of Sept. 6, 1967. The snow had finally disappeared in early July, and the summer was relatively dry. This time each of the 10 injections was made in separate trees to avoid the problem of two paths in one tree being confused due to spiraling ascent. The injections were again split evenly between $Na_2H^{32}PO_4$ and $Na_3{}^{32}PO_4$ using 5 mCi of ^{32}P per injection.

Results

The specific results of these initial tests were:

1. Water flowed slowly in red fir during midwinter under a snowpack 10 to 12 ft deep. Average rate of isotope ascent was 0.3 to 1.3 cm/hr during very unsettled weather conditions.

2. At the beginning of warm, clear weather in spring, rates of movement in both pine and fir were as great as 2 m/hr.

3. During summer, short-term rates of isotope ascent in the middle of the day ranged generally from 20 to 70 cm/hr. Nighttime rates were 2 to 7 cm/hr. No consistent differences between species were found.

4. The channel of ascent was narrow in both lodgepole pine and red fir, and the paths tended to spiral dextrally up the boles.

5. In the lower part of the boles, the isotope was concentrated in the outer 2 to 3 cm of sapwood, but some radioactivity was found as deep as 6 to 8 cm during periods of rapid ascent.

*Actually the hard ^{32}P beta radiation is converted to electromagnetic or bremsstrahlung radiation, and this is detected and measured by the G–M tube.

†For all but the last experiment, a hole that was large enough for at least two people had to be dug in the snow at each injection point.

6. At about 500 cm above ground, the paths usually began to spiral toward the center of the boles, and near the tops of the crowns, the paths had spread over most of the cross-sectional area. (RHL)

REFERENCES

References marked with an asterisk have not been verified by the editor; for further explanation, see the Foreword.

*1. R. E. Dills, Influence of Forest Cutting and Mountain Farming on Some Vegetation, Surface Soil, and Surface Run-Off Characteristics, U. S. Forest Service, Southeastern Forest Experiment Station, Station Paper No. 24, 55 pp., 1953.

*2. K. G. Reinhart, A. R. Eschner, and G. R. Trimble, Jr., Effect on Streamflow of Four Forest Practices in the Mountains of West Virginia, U. S. Forest Service Research Paper NE-1, Northeastern Forest Experiment Station, 79 pp., 1963.

*3. L. R. Rich, H. G. Raynolds, and J. A. West, The Workman Creek Experimental Watershed, U. S. Forest Service, Rocky Mountain Forest and Range Experiment Station, Station Paper No. 65, 18 pp., 1961.

*4. H. I. Penman, Vegetation and Hydrology, *Commonwealth Bur. Soils*, **53**: 124 (1963).

*5. C. W. Thornthwaite, An Approach Toward a Rational Classification of Climate. *Geograph. Rev.*, **38**: 55-94 (1948).

*6. E. R. Burroughs, Jr., and J. D. Schultz, Evapotranspiration and Soil-Moisture Depletion, *Proc. Soc. Amer. Foresters*, **1964**: 98-101 (1964).

7. A. J. West and K. R. Knoerr, Water Losses in the Sierra Nevada, *J. Amer. Water Works Ass.*, **51**: 481-488 (1959).

*8. R. R. Ziemer, Soil-Moisture Depletion Patterns Around Scattered Trees, U. S. Forest Service Research Note PSW-166, Pacific Southwest Forest and Range Experiment Station, 13 pp., 1968.

9. I. P. Arthur, An Evapotranspirometer, *Aust. J. Agr. Res.*, **6**: 707-712 (1955).

10. C. H. M. van Bavel, Lysimetric Measurements of Evapotranspiration Rates in the Eastern United States, *Soil Sci. Soc. Amer. Proc.*, **25**: 138-141 (1961).

11. J. P. Decker and C. M. Skau, Simultaneous Studies of Transpiration Rate and Sap Velocity in Trees, *Plant Physiol.*, **39**: 213-215 (1964).

*12. A. Halevy, Orange Leaf Transpiration Under Orchard Conditions, IV. A Contribution to the Methodology of Transpiration Measurement in Citrus Leaves, *Bull. Research Council Israel*, **5D**: 165-175 (1956).

*13. A. J. Rutter, Evaporation from a Plantation of Pinus sylvestris in Relation to Meteorological and Soil Conditions, *Publs. Ass. Int. Hydrol. Scient.*, **48**: 101-110 (1949).

14. K. N. H. Greenidge, Rates and Patterns of Moisture Movement in Trees, in *The Physiology of Forest Trees*, pp. 19-41, K. V. Thimann (Ed.), The Ronald Press Company, New York, 1958.

15. J. A. Rudinsky and J. P. Vité, Certain Ecological and Phylogenetic Aspects of the Pattern of Water Conduction in Conifers, *Forest Sci.*, **5**: 259-266 (1959).

16. T. T. Kozlowski, J. F. Hughes, and L. Leyton, Patterns of Water Movement in Dormant Gymnosperm Seedlings, *Biorheology*, **3**: 77-85 (1966).

*17. B. Hüber and E. Schmidt, Ein Kompensationsmethode zur thermoelektrischen Messung langsamer Saftstrome, *Ber. Deut. Bot. Ges.*, **55**: 514-529 (1937).

*18. R. H. Swanson, An Instrument for Detecting Sap Movement in Woody Plants, U. S. Forest Service, Rocky Mountain Forest and Range Experiment Station, Station Paper No. 68, 16 pp., 1962.

19. C. M. Skau and R. H. Swanson, An Improved Heat-Pulse Velocity Meter as an Indicator of Sap Speed and Transpiration, *J. Geophys. Res.*, **68**(16): 4743-4749 (1963).

*20. R. H. Swanson, Seasonal Course of Transpiration of Lodgepole Pine and Engelmann Spruce, in *International Symposium on Forest Hydrology*, W. E. Sopper and H. W. Lull (Eds.), pp. 417-433, Pergamon Press, Inc., Oxford, 1967.

*21. D. Doley and B. J. Grieve, Measurement of Sap Flow in a Eucalypt by Thermoelectric Methods, *Aust. Forest Res.*, **2**: 3-27 (1966).

*22. B. Hüber, Beobachtung und Messung pflanzlicher Saftstrome, *Ber. Deut. Bot. Ges.*, **50**: 89-109 (1932).

*23. W. K. Ferrell and E. E. Hubert, The Use of Radioisotopes in Forest Tree Research, *Idaho Forester*, **34**: 42-45 (1952).

24. D. A. Fraser and C. A. Mawson, Movement of Radioactive Isotopes in Yellow Birch and White Pine as Detected with a Portable Scintillation Counter, *Can. J. Bot.*, **31**: 324-333 (1953).

25. C. H. Beckman and J. E. Kuntz, Translocation of Poisons, Dyes, and Radioiodine, and Its Relation to Oak Wilt, (Abstract), *Phytopathology*, **41**: 2-3 (1951).

*26. V. G. Atrokhin, Radioautography in Forestry, *Lesn. Khoz.*, **11**(6): 15-19(1958). (Translation: PST Catalog No. 259, Office of Technical Services, U. S. Department of Commerce, OTS 60-21868).

27. S. N. Postlethwait and B. Rogers, Tracing the Path of the Transpiration Stream in Trees by the Use of Radioactive Isotopes, *Amer. J. Bot.*, **45**: 753-757 (1958).

28. F. J. Wray and J. A. Richardson, Paths of Water Transport in Higher Plants, *Nature*, **202**(4930): 415-416 (1964).

29. D. C. Lewis and R. H. Burgy, The Relationship Between Oak Tree Roots and Groundwater in Fractured Rock as Determined by Tritium Tracing, *J. Geophys. Res.*, **69**: 2579-2588 (1964).

30. F. W. Woods and D. O'Neal, Tritiated Water as a Tool for Ecological Field Studies, *Science*, **147**: 148-149 (1965).

31. D. F. Moreland, A Study of the Translocation of Radioactive Phosphorus in Loblolly Pine, *J. Elisha Mitchell Sci. Soc.*, **66**: 175-181 (1950).

*32. J. I. Farrar, Distribution of Radiophosphorus in Red Pine Seedlings, Canada Department of Resource and Development, Forestry Branch, Silviculture Leaflet 78, 1953.

33. A. I. Akhromeiko and M. V. Zhuravleva, Investigation of the Velocity of the Water Current in Woody Plants, *Soviet Plant Phys.*, **4**: 164-170 (1957).

*34. M. Klemm and W. Klemm, Die verwendung von Radioisotopen zur kontinuierlichen Bestimmung des Tagesverlaufes der Transpirationsstromgeschwindigkeit bei Baumen, *Flora*, **154**: 89-93 (1964).

35. C. A. Black, *Soil–Plant Relationships*, 332 pp., John Wiley & Sons, Inc., New York, 1957.

XUM

Old Bremsstrahlung University: Two Decades of Training in the Nuclear Sciences

By John F. Yegge* and Grover Smithwick*

Abstract: *Since its inception in 1948, Old Bremsstrahlung University (Special Training Division, Oak Ridge Associated Universities) has fulfilled a unique role in the area of training and education in the atomic and nuclear sciences. Over the years, expansion of the activities at the Special Training Division of Oak Ridge Associated Universites has paralleled the growth of the nuclear sciences in the fields of education, medicine, research, and technology, resulting in several particularly effective programs.*

It is an interesting exercise to consider requirements that might be imposed on an educational organization to give it the smallest probability of success. One might stipulate, for example, that no academic credit be granted by the institution for studies successfully completed or that very few courses be offered and that those few be conducted for at least 8 hr per day. One could certainly doom the institution to obscurity by giving it no campus and by having it hold classes in an abandoned document storehouse or in a vacant Army field hospital hurriedly constructed during wartime. The institution might even be situated on a fortified military base where public travel is forbidden by armed guards.

An "educational nightmare," created in the summer of 1948, had all these features. The first class of 32 intrepid students flippantly referred to the then unnamed Special Training Division of the Oak Ridge Institute of Nuclear Studies, now Oak Ridge Associated Universities (ORAU), as "Old Bremsstrahlung University." Little did anyone imagine that 20 years later, Basic Radioisotope Course No. 118 would be in session in that same "abandoned" storehouse.

The seed that was to bloom into the Special Training Division was planted only a month after the great Oak Ridge secret was revealed to the world at Hiroshima and Nagasaki. The occasion was a retirement party given in honor of Katherine Way, a member of the University of Tennessee Physics Department. Dr. Way commented to William G. Pollard (the first and present executive director of ORAU) how wonderful it would be if the scientific staffs of Southern universities could take advantage of the unparalleled research facilities in Oak Ridge. The result of ensuing discussions was an association of Southern universities (41 in number today) dedicated to spreading knowledge of this fantastic "new" science which had been made possible with the sudden availability of reactor-generated radioisotopes.

Then, as today, the training facilities were owned by the U. S. Atomic Energy Commission (or its precursor, the Manhattan Engineer District). Although all Division activities are conducted under contract with the Commission, other federal agencies such as the National Science Foundation (NSF) now often furnish partial support for specific courses.

SPECIAL TRAINING COURSES

Although the activities of ORAU special training courses have expanded into medical research, public education, and even job retraining, the Special Training Division continues to carry out the original responsibility of the association. The Radioisotope Techniques Course is designed to assist scientific, engineering, and technical personnel in obtaining a reasonable facility with radioisotope techniques as well as an insight into the safe and effective application of radioisotopes in their respective fields. Course content includes the theory of radioactivity, characterization of radiation, its interaction with matter, and systems for measuring radiation. Principles and practices of radioisotope applications are also presented. During the course, participants have an opportunity to use a great variety of detection systems and nuclear electronics. These range from relatively simple ionization chambers and Geiger counters to the more complex liquid scintillation spectrometers and multichannel analyzers. The principles that are learned during part of the course are applied to various situations such as activation analysis, isotope dilution analysis, and various radiochemical separation techniques later in the course.

The Medical Radioisotope Course is designed for physicians who need a knowledge of the basic principles of nuclear medicine. Practical experience with nuclear medical instrumentation and with various diagnostic tests and studies which may be performed

*Special Training Division, Oak Ridge Associated Universities; Mr. Yegge is on a leave of absence this year to do graduate work at Harvard University.

using radioisotopes is an integral part of this course. While this course contains some of the same information as the Basic Course, the emphasis throughout on the physician's use of radioisotopes requires special emphasis on radiation dosimetry and experience in medical radiation safety techniques and monitoring.

The 10-week course in Health Physics and Radiation Protection includes the material in the Basic Course, but with more thorough coverage of those areas pertinent to this field. In addition, this course consists in lectures and laboratory exercises on the problems and practices of providing radiation protection, radiobiology, radiation dosimetry, waste disposal, etc. Emphasis is placed on the methods and procedures necessary to evaluate radiation hazards and includes on-the-job training at both ORAU and Oak Ridge National Laboratory. The course is designed primarily for employees of state or local agencies having responsibility for radiation control. Others may be admitted through special arrangements.

The 4-week course in Nuclear Medical Technology is designed especially for practicing medical technologists, and the course content is quite similar to the Medical Radioisotope Course. However, greater emphasis is placed on nuclear medical instrumentation than on the techniques used in the various diagnostic procedures.

Special courses are run from time to time, usually of 3 weeks' duration, to present one of the topics of the above courses in more depth. Recent examples are the Neutron Activation Analysis Course and the Liquid Scintillation Counting Course. These courses are now being scheduled on a regular basis, and information about them can be obtained by writing to the Special Training Division. (See Fig. 1.)

Some course *names* are the same today as they were when the Division was organized, but there are few other similarities. Over the years, new content, new instrumentation, and new teaching methods have been introduced, developed, and incorporated into the courses. More than 4000 participants have successfully completed the Basic Techniques Course alone, which has been but the nucleus for the proliferation of other specialized courses (attended by another 4000 participants) in the burgeoning nuclear sciences; hundreds of colleges and universities across the nation have courses patterned directly or indirectly after their precursor in Oak Ridge.

The impact of courses, however, cannot always be measured by numbers alone. The total enrollment of such highly specialized offerings as Neutron Activation Analysis, Liquid Scintillation Counting, and Small Accelerator Physics, for example, is not great because they are offered infrequently and the potential audience is limited. But there is a need for such courses, and the policy of the Special Training Division has been to provide training not readily available in conventional university courses. When appropriate, experts from the field supplement the training provided by the Regular Division staff and help keep course content current.

Fig. 1 University professors and scientists from both the government and private industry participate in 4-week Radioisotopes in Research programs. The first 2 weeks stress the basic physics of radiation and radiation detection. During the last 2 weeks, studies in each participant's area of special interest are pursued.

Perhaps no characteristic is more uniform throughout the Special Training Division staff than the high level of instructional talent which has been developed. The combination of a staff which is conscious of the great importance of excellence in teaching with a large collection of modern radiation-detecting devices has catapulted the Division into a position of international educational prominence. Hundreds of laboratory experiments have been written and are widely copied; dozens of journal articles have been published; and texts as well as books of experiments and demonstrations have been produced—all by men who are as respected among educators as among scientists.

MOBILE RADIOISOTOPE LABORATORIES

As the demand for training in uses of radioactivity grew among college and university faculties, the facilities in Oak Ridge were not adequate to meet all the needs. The problem was compounded by the fact that most college instructors were able to attend training sessions away from home only in the summer.

XUM

In the Fall of 1959, the Special Training Division provided a solution to both these problems by constructing and staffing a van-mounted radioisotope training laboratory designed to be driven to college campuses for 2-week periods of intensive use by science faculties and, space permitting, advanced science students. In addition to the driver–laboratory instructor, two experts in radioisotope techniques each spend a week on the campus lecturing to the participants and, frequently, to fairly large groups of other interested persons. (See Fig. 2.)

Today three of these laboratories are crisscrossing the nation, having made more than 250 visits in 41 states and the District of Columbia and having reached more than 8000 participants. Two other mobile laboratories, also designed by the Special Training Division, are touring Asia and South America under the sponsorship of the International Atomic Energy Agency. An index of the astonishing impact of the mobile-laboratory program is that no promotional literature has ever been needed to maintain a 2-year backlog of requests for domestic visits.

INTERNATIONAL EXHIBITS

The efforts of the Special Training Division are also international in scope. The AEC's exhibit, "Atoms in Action," which tours Latin America, is heavily staffed

Fig. 2 Transistorized electronics are employed on the mobile radioisotope laboratory to obtain maximum use of the available space. The vans contain Geiger–Mueller counting systems and gamma-ray spectrometers in quantities that will accommodate eight participants at a time. In addition, there are three radiochemical fume hoods, a library and a large supply of replacement parts for on-the-spot servicing of equipment by the laboratory instructor. More than 250 colleges have been served with training being provided to over 8000 participants. This photograph was taken during a recent 3-week visit to the campus of Federal City College, the newly opened college for disadvantaged students in Washington, D. C.

by scientists from the Division. The responsibilities of these scientists range from the showing of secondary school teachers how to present exciting lecture–demonstration programs to the training of local scientists in modern research techniques. Special courses are presented for nuclear electronics engineers, physicians, university professors, research scientists, and teachers. The dedication of the staff to the job is reflected by the fact that many have mastered the Spanish language well enough to present all their lectures in the local tongue. In addition, four Spanish-language course manuals have been written and have become definitive works in the language. In preparation for the visit of "Atoms in Action" to São Paulo, Brazil, these same scientists prepared themselves to lecture in Portuguese. (See Fig. 3.)

The Special Training Division also has an important role in the "Atoms in Action" exhibit that tours Europe and Asia. The Division's main effort there is in the high school classroom part of the exhibit in which thousands of local high school students are shown how the peaceful use of atomic energy will make the world a better place in which to live. Teaching techniques and devices, conceived and developed in Oak Ridge, usually generate considerable enthusiasm among the local teachers.

RECENT ACHIEVEMENTS

Under the guidance of the present chairman, Lawrence K. Akers, the emphasis continues on the Division's objective of improved science education at all levels. A few examples of recent achivements will illustrate the broad spectrum of the success of these efforts:

Experiments in the use of low-energy Cockcroft–Walton particle accelerators for undergraduate physics instruction have been developed under a joint NSF–AEC

Fig. 3 Until recently the AEC's "Atoms in Action" exhibit in South America has been presented in this temporary inflated nylon structure. Future exhibits, however, will be conducted in three 80-ft inflated concrete domes that will be at the exhibit site for continuing use by the host country. The Special Training Division's role in the exhibit has resulted in the preparation of several Spanish language training manuals. These manuals have become the definitive Spanish language works on the subject.

XUM

grant; this project spawned six theses, a score of journal articles, and the Proceedings of the Conference on the Use of Small Accelerators for Teaching and Research* held at ORAU, April 8–10, 1968, and sponsored by the U. S. Atomic Energy Commission.

Cooperation between the Special Training Division and science educators of Harvard University culminated in the development of an appreciable part of a new and innovating high-school and junior-college physics curriculum—Project Physics. The Special Training Division has conducted two Project Physics institutes, among its 17 NSF–AEC sponsored summer institutes, for high-school and junior-college teachers and has recently presented the Project Physics curriculum to administrators and science educators at state-level meetings in Alabama, Tennessee, and Georgia.

Academic year institutes have been conducted for junior-college physical science teachers for 3 years since 1965; more than 30 master's degrees (granted by the University of Tennessee) have been earned in this way in addition to the several papers that have been presented at scientific meetings and journal articles that have been published.

Experiments in the use of isotopic neutron sources for activation analysis were developed for use in colleges; the resulting publication is presently in its fourth printing.†

A special course for nuclear medical technologists was developed; the demand for this offering required that it be presented three times in its present third year as compared with twice in the second year and once in the first year.

The radiocarbon dating laboratory continues to be internationally recognized as a standard against which other dating laboratories are judged.

The U. S. Office of Education has recently begun to support Division activities with a grant for the retraining of 2- and 4-year-college science teachers from developing and disadvantaged institutions. This 11-month program will enable several science instructors to earn masters' degrees through the University of Tennessee.

Apparent throughout the Division's activities is the unifying thread of concern for how its efforts can best be directed to the cause of improving science education at all levels. Even the original research, undertaken at the Special Training Division, has a distinctively educational flavor and, undoubtedly, partly accounts for the Division's continuing leadership in science education.

PLANS FOR THE FUTURE

One cannot help but sense the enthusiasm at the Special Training Division: scientists converse in their "new" language over lunch; laboratory instructors earnestly help eager science students from nearby (less than 600 miles distant) colleges who are in Oak Ridge for a week of radioactivity training; physicians test diagnostic procedures on dogs; teachers from Taiwan or Brazil diligently rehearse their lectures for "Atoms in Action." If concern for the future rather than the past is a sign of vitality, then the future is bright, indeed, for the Special Training Division. (See Fig. 4.)

Fig. 4 An exciting activity that the Division has recently begun to explore is very short and intensive radioactivity training courses for college students of nearby colleges. Students in advanced science classes spend up to 1 week in Oak Ridge supplementing the basic training they receive at their home institutions. Shown here with an instructor are two of a group of 30 science students representing 11 area colleges, each of which sent its most outstanding science students.

A committee of Division staff members is presently considering how the Division's efforts should be directed 5 years and more from now. Plans are being made, for example, on how to use the new ^{252}Cf neutron source most effectively. Present plans include its use as a research tool in activation analysis by college professors who would not ordinarily have access to this type of source and the necessary supporting electronic apparatus.

Another area receiving considerable thought is the use of computers for programmed study. While basic computer application use has already been incorporated in all Special Training Division courses, it is anticipated that this will be one area of major expansion due to its educational applications for both the professional and educational communities.

A 6-week course in nuclear criticality safety is being developed for its first presentation in 1970. It is planned to first meet the immediate need for training

*USAEC Report CONF-680411.
†USAEC Report ORAU-102.

practicing nuclear safety engineers. Then the long-range problem will be attacked by giving specialized training to college professors of nuclear engineering, hoping that they will transmit an attitude of safety consciousness to their students by having it pervade their entire instruction.

The Division will continue to create educational techniques and do research in areas where an educational need is detected. Although research is not immediately consistent with the educational orientation of the Division, its staff cannot help but be aware of the research possibilities uncovered in its training activities. For example, an educational experiment is being prepared for the Brazilian edition of the "Atoms in Action" exhibit in which radioisotope training is conducted, using individualized instruction. This program is multidimensional and includes programmed and computer-managed instruction.

The Division's prominence in secondary-school and junior-college science teacher training will undoubtedly continue—especially as the study of radioactivity and the nucleus finds increased emphasis at the introductory level. And methods developed in the Science Teacher Program will continue to be spread to other lands in the international "Atoms in Action" exhibit program.

Other, now unforeseen, educational programs and activities as well as education-oriented research will surely be a part of the future for Old Bremsstrahlung University. The fruition of enthusiasm, understanding, and talent developed during the past two decades will certainly continue to be prominent. (RHL)

Radioactive Tracer Technique for Laboratory Corrosion Studies

By Harry J. Close and D. B. Hines*

Abstract: *A radioactive tracer method for determining the corrosion rates of mild steel and 316 stainless steel in sulfuric acid at room temperature was developed. Irradiated coupons of the metal under test were exposed to sulfuric acid, which was circulated to a flow cell in the well of a scintillation counter. The corrosion rates were determined by comparing the counting rates with those from known weights of irradiated comparison standards.*

Since process development chemists and chemical engineers lack a rapid, reliable technique for continuous measurement of corrosion rate vs. time, a method was developed that uses a radioactive tracer technique for the continuous measurement of corrosion on test specimens. Although conventional methods of immersion testing (the amount of material brought into solution by corrosion is determined by standard analytical methods) have been used extensively, they do not always yield the desired sensitivity, speed, or specificity—i.e., capability of determining more than one component (for example, iron and chromium) of the material subject to corrosion. A literature search showed numerous applications of radioactive tracers to corrosion studies, usually involving autoradiographic techniques. But no reference dealt precisely with our proposed technique—use of a radioactive specimen and continuous measurement of the radioactivity accumulating in a corrosive solution. This technique is somewhat similar to that in which radioactive tracers are used in friction and wear studies, where wear of radioactive samples is evaluated by continuous measurement of the activity buildup in a lubricant.

For our work, a simple corrosion system of well-known rate and type of attack was chosen to permit simple, yet reliable, experimental design. The corrosion of mild steel and 316 stainless steel in sulfuric acid at room temperature was selected for study.

EXPERIMENTAL

The equipment used (Fig. 1) can be divided into two logical groupings: (1) equipment for exposing a test coupon to a corrosive solution and (2) apparatus for determining the radioactivity in the corrosive medium. The exposure equipment consisted of a constant-temperature bath containing two flasks of the agitated corrosive agent—one containing a radioactive coupon and the other a nonradioactive control. The

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corrosive solution in the flask containing the radioactive coupon was circulated through the activity-detection apparatus by a pump. The measuring and recording equipment consisted of a 65-ml flow cell in a well-type sodium iodide crystal scintillation detector (shielded with lead bricks), two single-channel gamma spectrometers, and two recording rate meters.

The nonradioactive control corrosion solution was sampled and analyzed for iron and chromium by an atomic absorption technique. The data from the radioactive solution were processed to obtain corrosion rate vs. time, as well as overall corrosion rate and percentage iron and chromium in the corrosive solution.

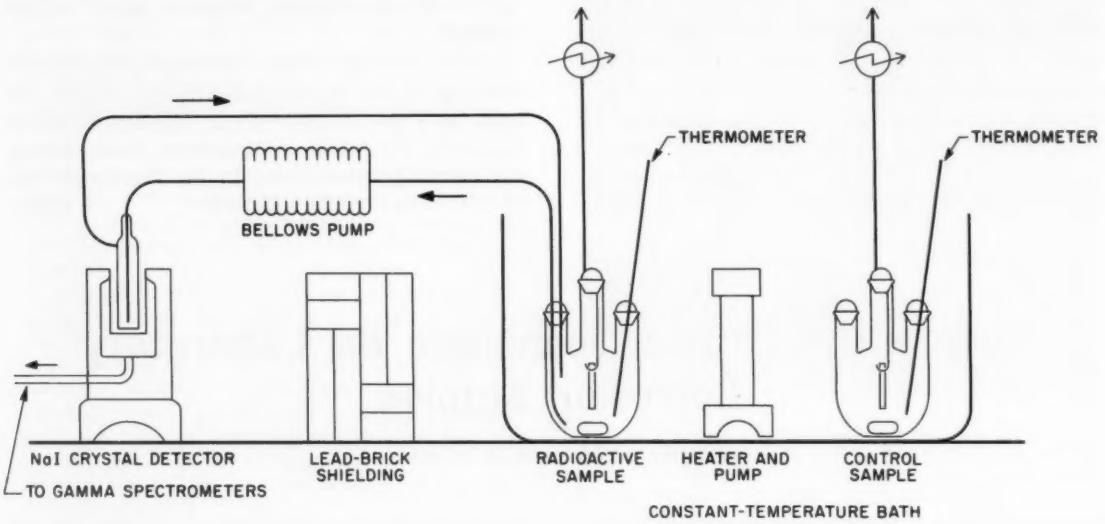

Fig. 1 Equipment diagram.

Corrosion coupons and metal turnings of both mild steel and 316 stainless steel were irradiated in a neutron flux at the University of Missouri to produce ^{59}Fe and ^{51}Cr isotopes. Comparison standards of both metals were prepared by dissolving known weights of radioactive metal turnings in known volumes of acid. Detector flow cells identical to the test cell were prepared and filled with the comparison standards for interpreting the test data.

Prior to immersion of the specimen in the corrosive solution, background and comparison standard counting rates were determined. A certified ^{60}Co radiation standard was used to determine instrument stability. The specimens were then immersed in the acid solution, and a run was started. Data, as counts per minute, were obtained continuously on rate-meter recorders and intermittently from digital readout counters or scalers.

At the end of each run, the system and coupons were thoroughly rinsed with water and dried, and the standards and background were recounted.

RESULTS

Corrosion of Mild Steel

Mild steel corrosion in 65-wt.% sulfuric acid was measured in four replicate runs and in a fifth run with 0.3-wt.% arsenic as sodium arsenite added as an inhibitor. All runs were made at 28°C for 5 to 6 hr.

The data from the first run (Fig. 2) agreed well with literature corrosion-rate values determined by conventional methods in immersion testing—about 20 mils/year. Data from the scaler and the recording rate meter agreed with each other.

In subsequent runs (Fig. 2) with the same coupon, corrosion rates were higher than in the first run. The higher rates are believed to be due to rusting between runs and to increased surface area resulting from corrosion during Run 1. The curves for Runs 2, 3, and 4 appeared to converge and to indicate a corrosion rate three to four times that of Run 1. These data show some correlation between downtime and initial rate of

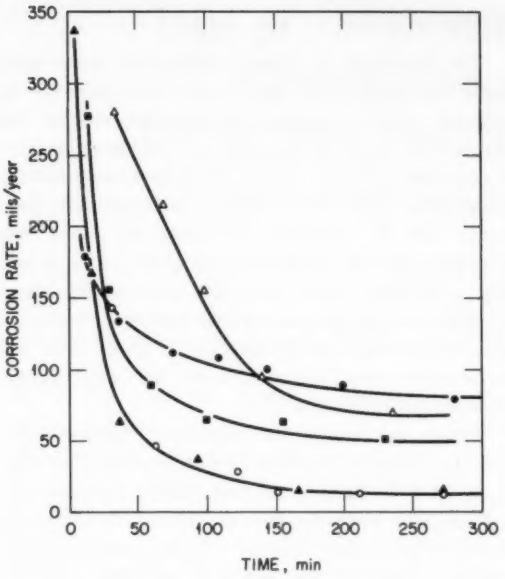

Fig. 2 Corrosion rates of mild steel in 65-wt.% H_2SO_4 at 28°C. ○, Run 1, data from scaler. ▲, Run 1, data from recorder. ●, Run 2, after 48 hr downtime. ■, Run 3, after 19 hr downtime. △, Run 4, after 90 hr downtime.

attack. Downtimes between Runs 1 and 2, 2 and 3, and 3 and 4 were 48, 19, and 90 hr, respectively.

Sodium arsenite was not very effective as an inhibitor as used in Run 5; however, the rate had not leveled out and might have decreased with more exposure time (Fig. 3). By the end of the run, the rate was 50 to 60% of the rates in Runs 2, 3, and 4.

Except for Run 1, agreement of test results from the radioactive solution and the control solution was as good as that expected for determination of corrosion rates in conventional immersion testing (Table 1). There is, of course, the possibility that surface damage may have resulted from irradiation, but more probably, the agitation in the two flasks was dissimilar.

Corrosion of Stainless Steel

Corrosion of 316 stainless steel in 65-wt.% sulfuric acid at 28°C was measured in two runs. A third run was made in 50-wt.% sulfuric acid at 28°C to which 0.7-wt.% ferric sulfate was added, not only to inhibit general corrosion but to accelerate intergranular attack. The first run with the radioactive tracer method showed a corrosion rate in close agreement with that found in the literature, determined by conventional

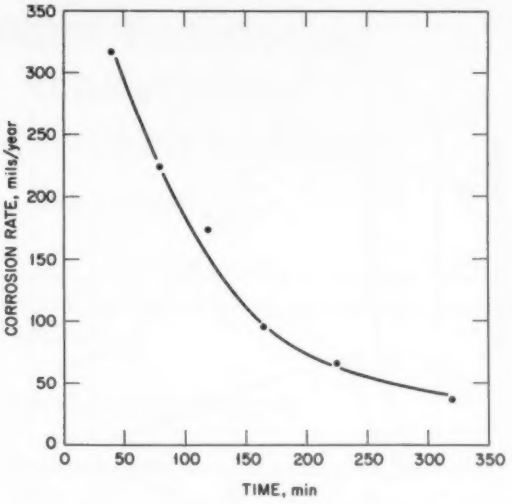

Fig. 3 Corrosion rates of mild steel in 65-wt.% H_2SO_4 at 28°C with 0.3-wt.% arsenic as $NaAsO_2$ (Run 5).

Table 1 Corrosion of Mild Steel in 65-wt.% H_2SO_4 at 28°C

Run No.	Metal in solution, ppM	
	Radioactive tracer technique	Atomic absorption technique
1	3.7	10.7
2	13.2	18.7
3	12.8	11.7
4	18.4	12.0
5	18.4	15.6

immersion testing methods—i.e., about 100 mils/year. Passive film breakdown was observed after about 20 min exposure. The corrosion rate reached a maximum at 65 min exposure, decreased slowly over the next 4 hr, and leveled off at about 100 mils/year (Fig. 4). Run 2 followed the same pattern as Run 1 except with respect to time. Passive film breakdown did not start until after 3 hr exposure and continued for the next 2 hr. The run was not continued long enough to determine whether or not the corrosion rate would have peaked and then leveled off at a lower rate. The longer time required to activate the metal surface resulted from air passivation between runs.

The objective of the third run with stainless steel was to detect intergranular attack by observing an

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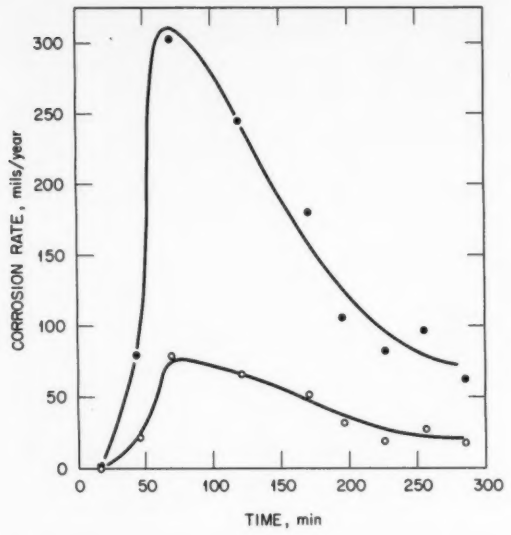

Fig. 4 Corrosion rates of 316 stainless steel in 65-wt.% H_2SO_4 at 28°C. ●, iron. ○, chromium.

iron/chromium ratio different from that of the bulk metal—i.e., 3.7/1. The sulfuric acid–ferric sulfate inhibits general corrosion but accelerates the chromium carbide type of intergranular attack. There was no evidence of selective attack—iron and chromium appeared in the corrosive solution in the same ratio as that of the alloy. Intergranular attack probably did not occur because the test was at room temperature rather than at higher temperatures and was of too short duration.

There was acceptable agreement between results from the radioactive tracer determinations and those from the control analyzed by atomic absorption (Table 2). The data do not indicate any surface damage from the irradiation.

Table 2 Corrosion of 316 Stainless Steel in H_2SO_4 at 28°C

Run No.	Metal in solution, ppM				
	Radioactive tracer technique			Atomic absorption technique	
	Fe	Cr	Fe : Cr	Fe	Cr
1	19.2	5.2	3.7	22.3	7.0
2	2.7	0.7	3.9	5.3	1.7
3	0.4	0.1	4.0	–	–

CONCLUSIONS

The feasibility of using a radioactive tracer technique for rapid—less than 8 hr—determination of corrosion was demonstrated in laboratory studies. The method has several advantages over the determination of corrosion rates by conventional immersion testing. Quantitative data, from which a corrosion rate–time profile can be obtained, are generated. Very high sensitivity can be achieved—corrosion rates of less than 1 mil/year were measured. High specificity is possible; for example, the determination of corrosion rates of individual components of the steel—iron and chromium—was demonstrated by dual-channel monitoring of ^{59}Fe and ^{51}Cr.

There are some limitations and disadvantages: (1) some metals and alloys do not have elements that form, upon neutron irradiation, usable radioisotopes that are gamma emitters with satisfactory half-lives and resolvable spectrums; (2) only one test coupon may be used per run; (3) the corrosive solution must be assumed to be a homogeneous system, and the corrosion products containing the radioactive isotope must be assumed to be uniformly mixed in the corrosive solution; (4) irradiation cost and instrumentation makes the radioactive tracer technique more expensive than conventional testing; and (5) special precautions must be taken to minimize the hazards connected with handling radioactive materials.

Corrosion testing with smaller specimens irradiated to higher levels could increase the sensitivity of the method, while at the same time reducing total radiation and exposure hazards.

Cobalt-60 might be used as a universal tracer to monitor corrosion of essentially nonferrous metals and alloys. Trace amounts of cobalt, which has a high thermal-neutron cross section, would provide sufficient activity for materials that cannot in themselves become gamma emitters. (REG)

BIBLIOGRAPHY

A. L. Glass, Recent Radioactive Tracer Method Measures Leaching Rate of Chromium, *Mater. Protect.*, 7(7): 26-30 (July 1968).

B. E. Gordon, P. J. Raifsnider, and D. L. Lilly, Corrosion Monitoring in Oil Wells with a Radioactive Specimen, *Mater. Protect.*, 5(11): 21-23 (November 1966).

C. D. Bopp and O. Sisman, How to Calculate Gamma Radiation Induced in Reactor Materials, *Nucleonics*, 14(1): 46-50 (January 1956).

A. Alon, M. Schor, and A. Vromen, Corrosion Resistance of Ta to Mixture of Phosphoric Acid and Potassium Chloride at 120°C, *Corrosion*, 22(1): 1-6 (January 1966).

Use of ^{252}Cf for Mineral Exploration: Comparison with Accelerators for In Situ Neutron Activation of Silver*

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Abstract: *Tests were made on the use of ^{252}Cf and accelerators as neutron sources for in situ neutron activation of silver, with gamma-spectrometer detection of the activated silver. Comparison of the results indicated some advantages of ^{252}Cf.*

For several years the federal government and the mining industry in the United States have been concerned with the increased consumption of silver. We therefore undertook a study to determine the feasibility of using in situ neutron activation in prospecting for new silver deposits. Early experiments[1,2] made with conventional isotopic neutron sources—e.g., ^{226}Ra–Be or ^{241}Am–Be—showed that, for the most easily activated elements, a flux of at least 10^8 n/sec is needed. A conventional isotopic neutron source of this yield requires a heavy gamma and neutron shield. A positive-ion accelerator provides a higher neutron flux and can be operated remotely, obviating the need for a shield. We used an accelerator for most of our work up until mid-1968, at which time a ^{252}Cf fission-neutron source suitable for field work was made available by AEC's DuPont-operated Savannah River Plant. This new source had a total neutron emission considerably in excess of that obtainable with a 150-kV accelerator using a deuterium target, the average energy was lower, and a minimum amount of moderator was required to decrease the neutron energy to thermal levels. Because of the relatively low gamma emission, a large gamma shield is not needed. Of course, a substantial neutron shield is required, but this, although it creates a handling problem, can be tolerated. One of the most attractive features of a ^{252}Cf neutron source is its small size and hence its potential use for mineral exploration in a borehole. Preliminary feasibility tests were therefore made with a ^{252}Cf neutron source in place of an accelerator for in situ neutron activation of silver ores. This report compares the data taken with the two sources in order to evaluate the use of ^{252}Cf as a neutron source for mineral exploration by in situ neutron activation techniques.

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SOURCE DESCRIPTIONS

Accelerator Units

For field operation the accelerator was mounted on vehicles (Fig. 1). The accelerator and a NaI(Tl) detector were separated by a composite paraffin–lead–cadmium shield and were mounted rigidly with respect to one another on a metal frame. The frame, actuated by a hydraulic mechanism, shuttled back and forth on wheels over a track on the vehicle tailgate. For irradiation, the tailgate was lowered, and the accelerator was placed in the center, over a large hole in the tailgate. At the end of the irradiation, the frame was moved to locate the detector over the irradiated spot, which could be done in as short a time as 3 sec. The accelerator used a deuterium target and produced about 10^8 n/sec at about 2.95 MeV. A boxed paraffin shield, between the vehicle and the accelerator, permitted the operator to remain safely in the vehicle during the irradiation.

In addition to the 150-kV accelerator, a 200-kV accelerator was also mounted on a truck (Fig. 2) for studying a wide variety of reactions and techniques. In this case the accelerator and tailgate automatically rose at the end of the irradiation. The target assembly was of the multiple-rotary type so that any one of three tritium or two deuterium targets could be used. Neutron yields were several orders of magnitude higher than those of the smaller accelerator. Operation was completely automatic, and, since no personnel were in the vehicles, a shield was not needed.

The gamma spectrometry was done with conventional 2- by 2-in. NaI(Tl) detectors and multichannel analyzers. Some work was also done with Ge(Li) detectors, but their efficiency was too low for the desired sensitivity. The methods of using these vehicles for mineral exploration have been described elsewhere.[3-5]

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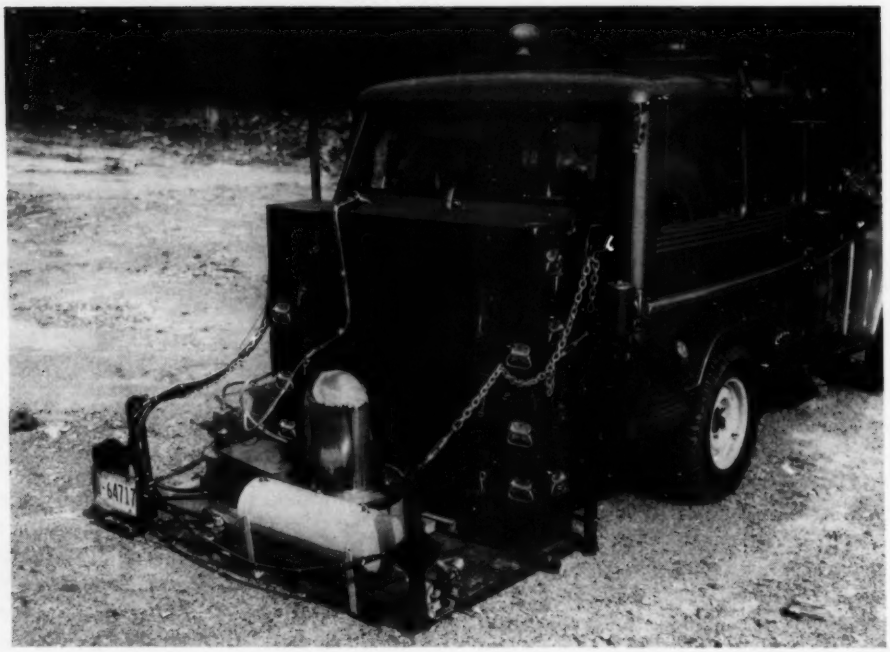

Fig. 1 Mobile silver exploration system. The 3-MeV neutron generator and detector are fixed to a shuttle, which moves across the tailgate, shown in irradiation position.

Fig. 2 Mobile system for silver exploration using 200-kV positive-ion accelerator; the 2- by 2-in. NaI(Tl) detector is mounted on a boom.

^{252}Cf Unit

The spontaneously fissioning neutron source, consisting of 90 μg of ^{252}Cf as the oxide in a platinum pellet, was doubly encapsulated in two small stainless-steel cylinders. This source assembly was mounted in a plastic insert, which fit into a hollow stainless-steel source holder (Fig. 3). The void volume in the source holder was nearly filled with oil to aid in scattering neutrons downward. The upper end of the source holder was attached to a 3-ft-long plastic rod for safe handling. The source yield was 2×10^8 n/sec.

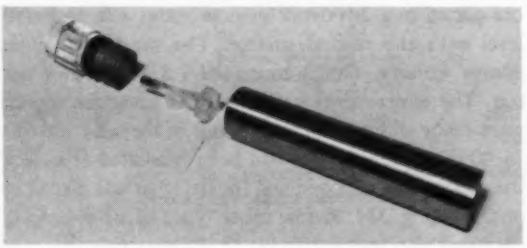

Fig. 3 The ^{252}Cf source and holder. The ^{252}Cf source pellet is mounted on the end of the hexagonal-head plastic screw.

A 55-gal drum of water with a 1.5-in. pipe welded in an axial position was used as a shield. The source holder was placed in the iron pipe and could be raised or lowered through the drum with the plastic rod. For in situ surface irradiation, the shield and source were mounted on the lift gate of a rugged four-wheel-drive vehicle (Fig. 4). For irradiation, the assembly was lowered and the source was dropped to the surface of the ground through a hole in the tailgate.

A few simulated borehole experiments were also made in the laboratory by placing several bags of ore around the hole on the top of the shielded drum. The source, plastic rod, and detector were interconnected with a single cable in such a way that the source could be raised out of the drum to a position in the center of the ore bags. At the end of the irradiation, the cable was lowered so as to place the detector in the former position of the source and the source in a shielded cavity below the drum.

Fig. 4 Shield (55-gal drum filled with water) for ^{252}Cf source in irradiation position on tailgate of vehicle.

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A 2- by 4-in. or a 3- by 3-in. NaI(Tl) scintillation counter and a multichannel analyzer with digital printout or $x-y$ plotting were used for the spectrum determinations. The counting equipment was mounted in a second vehicle to decrease exposure to both the operator and the crystal. At the end of an irradiation, the tailgate and source were raised, and the source vehicle was moved about 100 ft away. At the same time, the detector vehicle was moved and the detector was placed over the irradiated spot. The minimum delay between the end of the irradiation and the start of the counting was 10 to 15 sec. Tests with this rather crude preliminary arrangement could not be used for radioisotopes with a half-life of less than about 7 sec.

For most of the work, the multichannel analyzer was preset in the "add" mode. The add count was followed immediately by a count for the same length of time in the subtract mode. The resulting data yielded a differential-count spectrum. Counting statistics with this technique were generally poor, but the irradiation-count cycle could be repeated several times to improve the reliability of the results.

RESULTS

For practical mineral exploration, irradiation times must be less than 10 min. Because of the short irradiation times, the two most useful isotopes of silver are ^{108}Ag and ^{110}Ag, with half-lives of 2.3 min and 24.5 sec, respectively. Silver-108 has prominent gamma transitions at 0.42 and 0.63 MeV and ^{110}Ag at 0.662 MeV. The 0.42-MeV gamma of ^{108}Ag is often heavily masked by radiation from interfering elements, and hence we use the 0.63- and 0.66-MeV peaks to look for the presence of silver.

The ratio of the ^{110}Ag peak to the ^{108}Ag peak after a 10-min irradiation is about 2 to 1. However, this irradiation time produces a substantial number of short-lived activities that tend to mask the ^{110}Ag peak. We have obtained good results by irradiating for about five half-lives of ^{110}Ag (125 sec) and counting for about one half-life (24 sec). Although much of the activity is lost in this technique, the ^{110}Ag peak suffers less interference from the longer lived activities that tend to mask it when longer counting times are used. To further decrease interference from other long-lived activities and accentuate the ^{110}Ag peak, the irradiation time in some cases was also shortened. Generally, the irradiation and counting times were adjusted to obtain the optimum ^{110}Ag peak, and at the same time the ^{108}Ag peak was used to confirm the silver signature. No attempt was made to fix these parameters, which would have to be done for a quantitative technique.

To test these techniques with each type of source, in situ irradiations were made in the field over several different types of silver ores buried at shallow depths in the ground. As the tests were made at different times and places, the simulated ore bodies were not exactly the same for all the experiments. The accelerator experiments were made over 500 to 600 lb of silver ore placed in a 2-ft-diameter hole about 1 ft deep and level with the natural surface. The ore (Zaca Mine, Alpine County, Calif.) contained 1.5 oz of silver per ton. The experiments with the ^{252}Cf neutron source were made with several buried bags of the same ore (20 to 40 lb). After the ore had been irradiated (Fig. 5*a*), the source was removed and the detector was placed in position (Fig. 5*b*). A few other types of silver-bearing ores were also tested with the ^{252}Cf source.

3-MeV Accelerator Source

The initial tests were made with an accelerator as a source of 14-MeV neutrons. The background due to fast-neutron reactions was so high that the silver peaks were completely masked.[3] Further accelerator studies were therefore made with the 200-kV accelerator using a deuterium target for production of 3-MeV neutrons and a swinging detector boom (Fig. 2). The irradiations were made for 75 sec in a total flux of about 5×10^8 n/sec using a hydrogenous reflector around the target assembly. After a 6-sec delay to move the accelerator away and the detector into position, a 24-sec count in the add mode followed by a 24-sec count in the subtract mode was made. In the resulting spectrum (Fig. 6) both the 0.63- and the 0.66-MeV peaks are evident. The identities of these peaks were further confirmed by checking the half-lives.

It should be noted that the counting statistics of the spectrum as a whole are not adequate for identification of the other peaks without further computer data processing. However, based on spectrographic analysis of the ore and a table of energies, we have tentatively labeled some of the peaks as possible short-lived activities. The 0.66-MeV peak probably has small ^{137m}Ba and ^{91m}Mo components due to part-per-million quantities of barium and molybdenum in the ore. The peak at 0.51 MeV is the annihilation peak due to one or more positron emitters in the ore—e.g., zinc or copper.

The lower parts of the accelerator and tailgate assembly became quite radioactive, which caused some

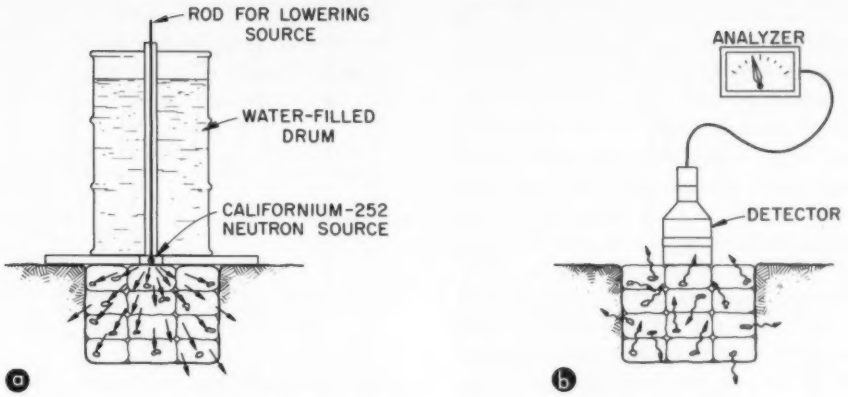

Fig. 5 (a) Californium-252 source and shield in irradiation position over buried ore bags, and (b) detector in position after irradiation.

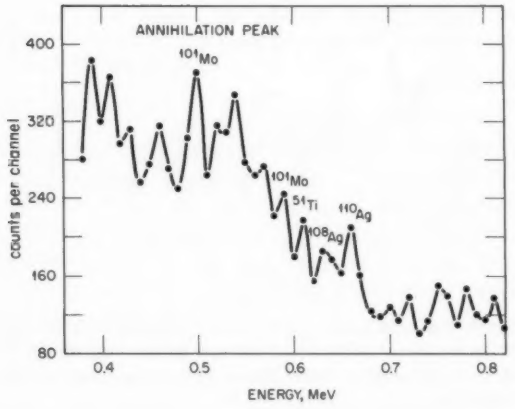

Fig. 6 Spectrum taken over silver ore (1.5 oz/ton) after 75-sec irradiation with accelerator 3-MeV neutrons. NaI(Tl) crystal (2 by 2 in.) used in 24-sec add–subtract mode count. Delay time ~10 sec.

interference even though the accelerator was moved 2 ft away. Interference was partially eliminated by shielding the NaI(Tl) crystal, but weight limitations on the boom precluded use of an adequate shield.

To improve the counting statistics, which are inherently bad for short counting times, a series of four successive cycles was made, each comprising a 75-sec irradiation followed by a 24-sec add–subtract counting mode. The cumulative data (Fig. 7) were considerably better even though no reflector was used behind the target assembly. In this case the 2.3-min ^{108}Ag activity is higher than that of the 24.5-sec ^{110}Ag. The cyclic irradiation and count were made with about 1 min between the sequences, and there was a buildup of the longer lived activity. In addition, the cumulative effect of this cyclic irradiation also brought out the 0.42-MeV peak of ^{108}Ag. For comparison, a silver quarter (1935) was placed on the ground and irradiated for 75 sec (24-sec count). Here, of course, the 0.66-MeV peak is larger than the 0.63, which appears merely as a point of inflection. By using the cyclic technique, it is evident that one could readily see about a tenth of the observed peak height, i.e., one could detect silver down to 0.1 or 0.2 oz/ton.

^{252}Cf Source

Figure 8 shows a spectrum taken with a ^{252}Cf source over about 35 lb of the same ore used in the accelerator experiments. Data recording was started within about 10 sec after a 125-sec irradiation. A 24-sec add–subtract mode count was made, and thus decay of the short-lived isotopes is chiefly represented. Both the 2.3-min ^{108}Ag and the ^{110}Ag activities are clearly visible. As before, the peaks were identified from the half-lives, and the other peaks in the spectrum are only tentatively assigned on the basis of energy and known spectrographic analysis. Although the flux from the ^{252}Cf source may be several times lower than that from the accelerator and the amount of ore irradiated is less, the results for a single irradiation appear to be better. To a large extent this is due to the larger fraction of low-energy neutrons initially emitted by the ^{252}Cf source compared with those initially emitted from an accelerator.

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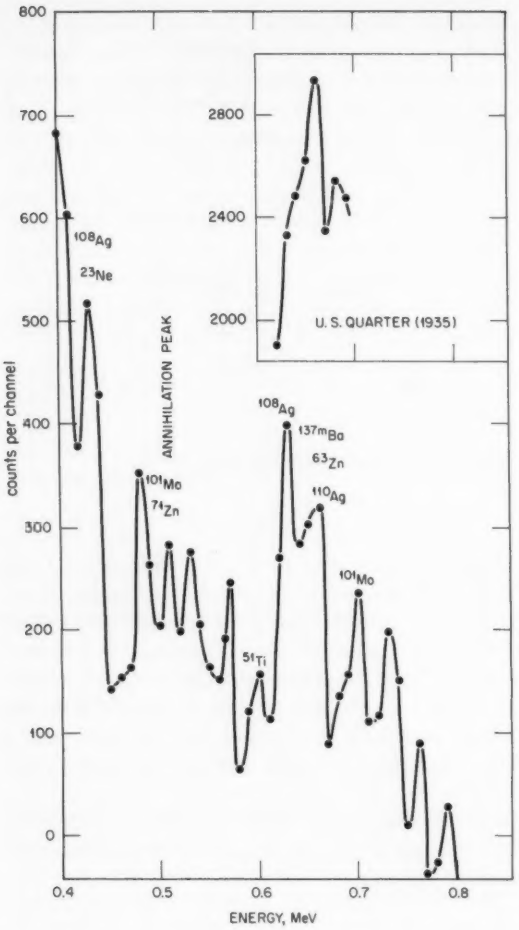

Fig. 7 Cumulative spectrum taken over silver ore (1.5 oz/ton) after four cycles comprising 75-sec irradiation with 3-MeV neutrons. NaI(Tl) crystal (2 by 2 in.) used in 24-sec add–subtract mode count. Insert shows, for comparison, the activity in a U. S. 1935 quarter (silver coin) under similar conditions.

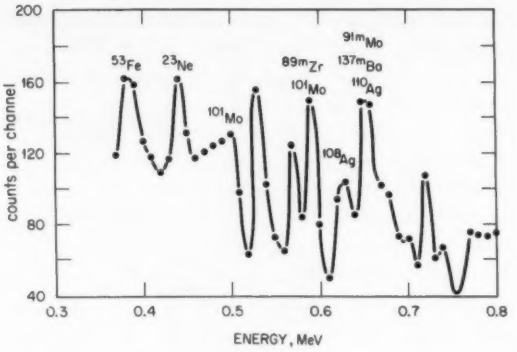

Fig. 8 Spectrum taken over silver ore (1.5 oz/ton) after 125-sec irradiation with ^{252}Cf neutron source. NaI(Tl) crystal (2 by 4 in.) used in 24-sec add–subtract mode count. Delay time 10 sec.

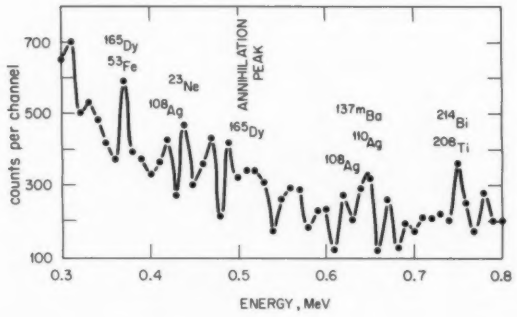

Fig. 9 Spectrum taken over silver ore (0.22 oz/ton) after cyclic 6-min irradiation with ^{252}Cf neutron source. NaI(Tl) crystal (3 by 3 in.) used in 1-min add–subtract mode count. Delay time 10 sec.

Although we did not try to carry out a cyclic irradiation in the above-mentioned experiment, one could improve the statistics of results by simply cumulating data from repeated runs. However, in examining a Florida phosphorite ore containing the rare earths, we used a cyclic 6-min irradiation with the ^{252}Cf source to detect dysprosium. Figure 9 shows the spectrum taken over about 4 lb of this ore, which contained 0.22 oz of silver per ton, using a 6-sec add–subtract mode count. Besides the ^{165}Dy peak at 0.37 MeV, peaks are also shown at energies close to the ^{108}Ag and ^{110}Ag peaks. The location of the larger of the two peaks in a position where the ^{110}Ag peak is expected is compatible with the relatively long irradiation time. The activities of both silver isotopes should be saturated at the end of the irradiation, and the ^{110}Ag/^{108}Ag activity ratio should be about 2/1. Although, at the time, silver was not known to be present in this ore, the peaks at the ^{108}Ag and ^{110}Ag positions and a ^{108}Ag peak at 0.42 MeV indicated the presence of silver. Subsequent spectrographic analysis of the ore showed a silver concentration of 0.22 oz/ton. The sensitivity with the ^{252}Cf source for silver exploration appears better than that of the accelerator using comparable techniques.

A relatively long irradiation was also made over native sandy soil in the experimental area, which was

thought to be barren of silver. After a 1-hr irradiation and 10-sec delay, a 24-sec add–subtract mode count was made. The differential spectrum showed a peak at 0.63 MeV and a smaller one at 0.66 MeV (Fig. 10).

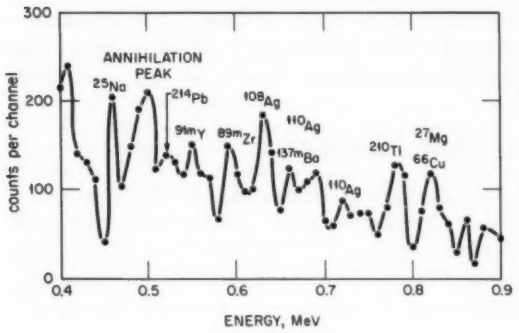

Fig. 10 Spectrum taken over barren sandy soil at the Savannah River Plant after a 1-hr irradiation with ^{252}Cf source. NaI(Tl) crystal (2 by 4 in.) used in 24-sec add–subtract mode count. Delay time 10 sec.

These peaks suggest the presence of silver, but half-life measurements of the peak in the ^{108}Ag position did not show a 2.3-min decay time. It was therefore interpreted as being due to other isotopes. However, spectrums made at the same spot several days later showed small peaks corresponding to the major transitions in the long-lived (253-day) ^{110m}Ag. Subsequent spectrographic analysis showed about 0.01 oz of silver per ton, which is probably the lower limit of detection with this particular ^{252}Cf source. The extended irradiation and delay times emphasize the gamma transitions of the long-lived ^{110m}Ag, and hence the use of this isotope would probably provide a more sensitive technique for determining the presence of extremely small amounts of silver. Because of the long irradiation time required, this technique of looking for silver would not be practical, although it illustrates what is probably the ultimate practical sensitivity.

To simulate the use of a ^{252}Cf neutron source for borehole exploration, several bags (35 lb) of the Zaca mine ore were placed around the source hole on top of the drum. With the source and detector on the same cable, the source was raised by remote control so as to be just out of the drum and in the center of the cluster of ore bags. After a 2-min irradiation, the source was dropped and the detector was placed in essentially the same position in about 2 sec. The better geometry of the simulated borehole allows higher counting rates, and the spectrum taken after a 2-min add–subtract count sequence shows both silver peaks (Fig. 11). However, unlike previous spectrums, the peaks here are about the same size. For the irradiation and delay times used, the ^{110}Ag peak would be expected to be nearly at saturation and hence higher than the ^{108}Ag peak. The reason for this was not at first obvious, but the slow-neutron flux of nearly 200 n/(cm^2)(sec), measured with a hand monitor at the position of the crystal during the counting, indicates an interference from prompt gamma emission. The prompt capture-gamma spectrum of thallium has a prominent 0.63-MeV peak, and we think that the enhancement of the ^{108}Ag 0.63-MeV peak was due to prompt gamma emission from thallium in the NaI(Tl) crystal. In an actual borehole this would not be a problem since the source–detector separation would be greater than was used in this experiment. The physical size and flux of the ^{252}Cf source appears to be nearly ideal for borehole work. As in the previous figures, the assignment of the other peaks is only tentative.

The silver cross-section curve has a number of resonance peaks in the epithermal region, and it was at first thought that epithermal neutron activation might enhance the silver peaks by decreasing interference due to thermal-neutron reactions. Silver-109 has a particularly large resonance peak, i.e., 10,000 barns. Spectrums taken with and without the ^{252}Cf source wrapped in cadmium showed little enhancement of the ^{108}Ag peaks and a substantial decrease in the ^{110}Ag

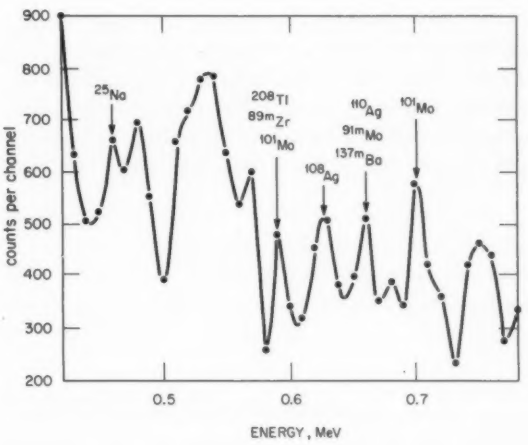

Fig. 11 Spectrum taken over silver ore (1.5 oz/ton) after a 2-min irradiation with ^{252}Cf source in simulated borehole configuration. NaI(Tl) crystal (2 by 2 in.) used in 2-min add–subtract mode count. Delay time 2 sec.

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peak. The large resonance peak in the ^{109}Ag cross-section curve applies primarily to the $^{109}Ag(n,\gamma)^{110m}Ag$ reaction, and thus the resonance integral for the $^{109}Ag(n,\gamma)^{110}Ag$ reaction is comparatively low.

COMPARISON OF SOURCES

Except when a pulsed-neutron application is essential, the ^{252}Cf source is to be preferred to an accelerator source of neutrons for a mobile operation, away from a laboratory. The most important advantages of the ^{252}Cf source are its small physical size and the minimum maintenance it requires. Accelerators have been made small enough for borehole use, but their flux is too low for their general use in a mineral-exploration device. The greatest practical problem with a ^{252}Cf source is the shield, but with proper construction this can be decreased to 250 to 300 lb, a tolerable weight for a practical field operation. With the accelerator, a large hydrogenous reflector around the target is desirable. However, an automatically programmed operation obviates the need for a formal shield.

Over short times, i.e., a few weeks, the flux from a ^{252}Cf source can be considered essentially constant, an important consideration for quantitative analysis. The neutron flux from an accelerator, on the other hand, requires constant monitoring and can seldom be reproduced from run to run. However, the accelerator can be turned off at will, while the ^{252}Cf source cannot.

The ^{252}Cf source emits a relatively small number of thermal neutrons and has an average energy of 2.3 MeV with the energy distribution peak at about 1 MeV. Hence, compared to the relatively high initial energy of accelerator-produced neutrons, a significant portion of the neutrons emitted from ^{252}Cf is initially at low energies and a minimum amount of moderator is required. A greater flux can be obtained through the part of the sample close to the point of detection than with a high-energy flux, which tends to distribute itself throughout the sample. On the other hand, accelerators can produce higher energy neutrons, which may be useful in observing a few fast-neutron reactions and for looking at a larger sample, whose depths can be penetrated. (MG)

ACKNOWLEDGMENTS

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REFERENCES

1. F. E. Senftle and A. F. Hoyte, Mineral Exploration and Soil Analysis Using In Situ Neutron Activation, *Nucl. Instrum. Methods*, **42**: 93-103 (1966).
2. A. P. Bushkov and V. I. Prokopchik, Determination of Fluorite by Activation Analysis, *At. Energ. (USSR)*, **21**: 215-217 (1966).
3. A. F. Hoyte, P. Martinez, and F. E. Senftle, Neutron Activation Method for Silver Exploration, *Trans. Met. Soc. AIME*, **238**: 94-101 (1967).
4. F. E. Senftle, Neutron Activation Techniques for Precious Metal Exploration, Final Report, USAEC Report TID-25169, 1967.
5. F. E. Senftle, P. Sarigianis, and P. W. Philbin, Neutron Activation Technique for Precious Metal Exploration, Can. Cent. Conf. Min. and Groundwater Geophys., Geol. Soc. Can., Econ. Geol. Series (in press).

Applications and Potential Applications of Radioisotopes to Melt-Shop Steelmaking Operations

By W. B. Tucker*

Abstract: *After providing background material to explain the various needs that radioisotopes can help meet in modern steelmaking melt-shop operation, this article reviews current uses and suggests some possible future uses of radioisotopes. Present uses include analysis of raw materials; identification of alloy elements; level gages; refractory wear gages; investigations of steelmaking kinetics; tracing sources of foreign inclusions; measurement of currents in the molten metal; and quick chemical analyses. Applications to casting include determination of solidification rate and location of liquid center in strand castings.*

Steelmaking, one of the first industries to apply radioisotopes, has recorded a large number of successful applications in both production gaging and research. Although some firms now depend heavily on radioisotopes for process control, indications are that the use of radioisotopes still meets unwarranted resistance, probably because of an information gap. Hence the advantages of their use need to be reviewed, although much has been written about radioisotope applications to steelmaking.

The subject also needs fresh exposure because of the rapid changes in processes, equipment, and methods presently taking place in the industry. Tremendous investments in new equipment are being made by all steelmakers. This new equipment is faster, has greater output, and operates to much more exacting specifications than the equipment it replaces. In a number of applications, radioisotopes do the job best. For a number of difficult design questions, radioisotope tracers can provide answers. Therefore modern managers and equipment designers should thoroughly know and exploit this technology.

This report is the first of a series of articles that will present an up-to-date picture of the widespread use of radioisotopes in steelmaking and suggest directions for future applications. Since these articles are directed toward people who understand steelmaking, the emphasis will be on today's and tomorrow's steelmaking methods. There will be no cataloging of the past applications because that has been adequately done.[1-4] This particular report will cover only melt-shop operations. Subsequent articles will cover the other phases of steelmaking. Of course, some of the applications mentioned here can also be used in other operations. The first section presents some background material that should help the average reader to better understand the significance of the application.

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THE MELT SHOP

Steel comes in thousands of standard shapes and sizes, and in each of these products there is the choice of one of a great number of standard chemical specifications or "grades." The steel buyer also has a great many options as to the crystal and grain structure, i.e., "qualities." Republic Steel Corporation alone produced more than a quarter-million combinations of bar grade and quality in 1969. When the complexity of the process equipment used to make such a full range of products available is considered, steel's low price is very remarkable.

The low cost of steel is the result of high-volume production methods with very specialized equipment, each item of which performs only a small step in the total process from raw material to finished steel. At the very center of production stands the melt shop. There the chemistry of the molten steel is adjusted to obtain a specific grade and a sound cast structure. Every part of steelmaking is important since every step must be performed correctly to give finished products of the highest quality; but the melt shop is of paramount importance because no subsequent step can convert poor starting material into high-grade finished products. Hence this is where any discussion of the application of radioisotopes to modern steelmaking must begin.

PROCESSES AND MATERIALS

This section describes the materials and processes used in modern steelmaking. Because of the rapidity of evolution of steelmaking, the definition of modern melt-shop practice (Fig. 1) given in the next few paragraphs may not satisfy everyone.

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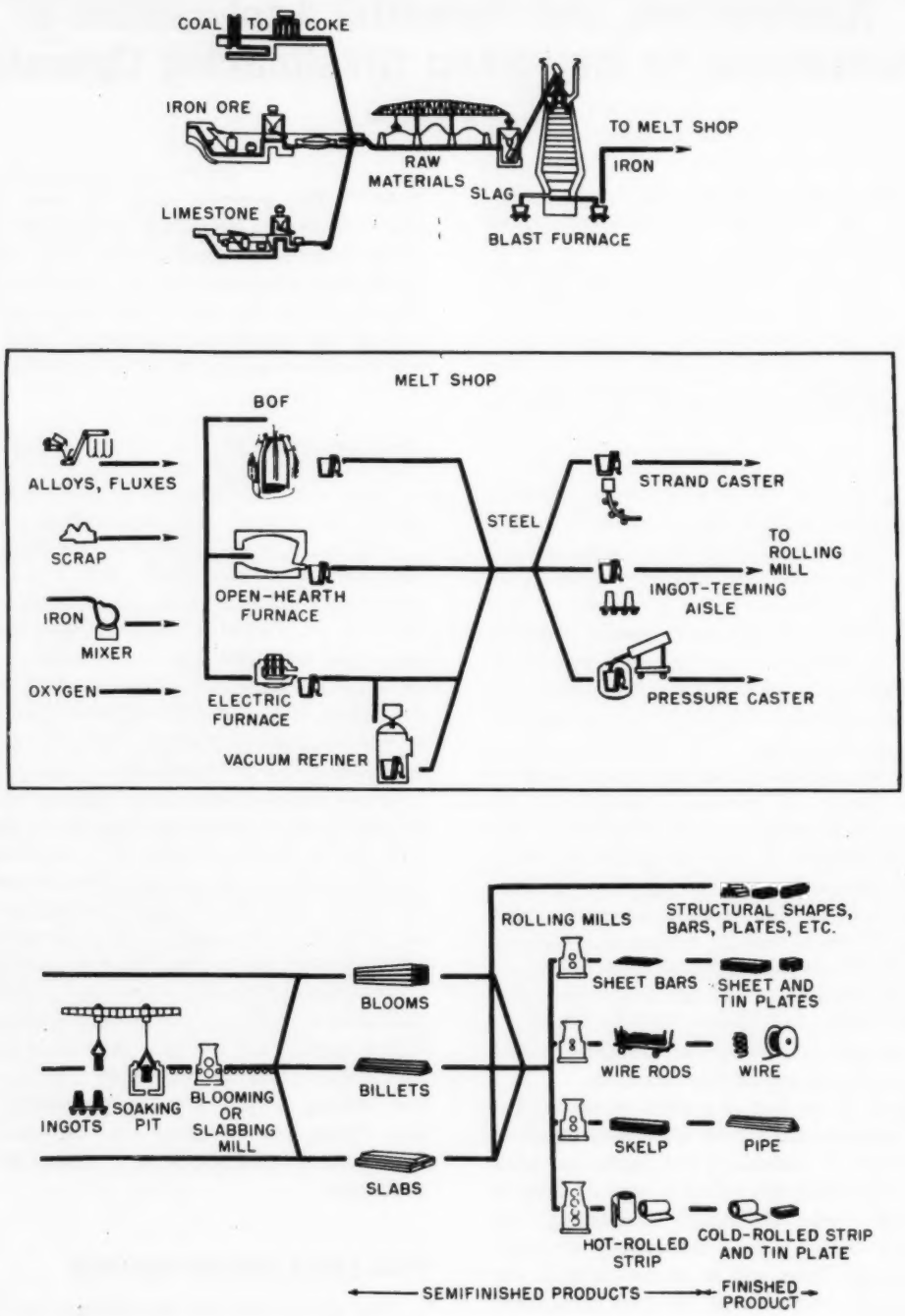

Fig. 1 Flow chart of steelmaking.

Iron, The Basic Raw Material

Iron, the primary raw material in steel, may come to the melt shop in the form of liquid hot metal from a blast furnace or in the form of solid scrap. The blast furnace is usually some distance away, necessitating transportation of the hot metal to the melt shop by rail in refractory-lined bottle cars. At the melt shop, it may be poured into a large vessel ("mixer") where the iron is mixed to maintain uniform composition and held until needed. The steelmaking furnaces are charged by pouring iron from a transfer ladle that is carried to the furnace (Fig. 2).

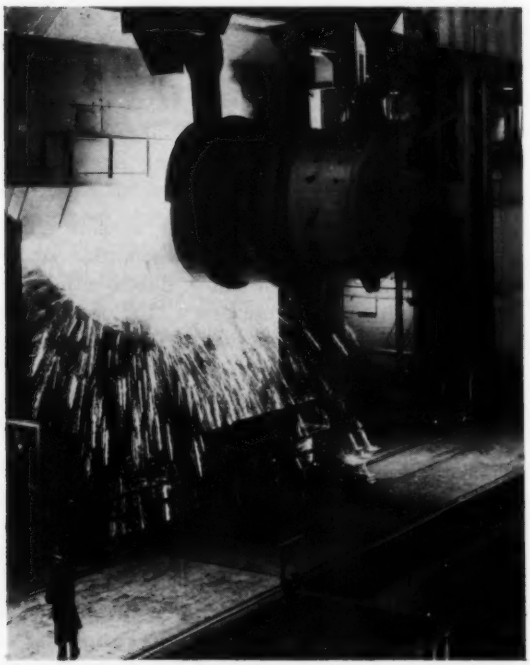

Fig. 2 A basic oxygen furnace (BOF) is charged with molten blast-furnace iron from a crane-held transfer ladle.

Scrap

Scrap is usually stockpiled on the ground at some distance from the melt shop, in piles of various grades. Railcar loads of the various grades are sent to a stocking area adjacent to the melt shop where stockers load scrap buckets with weighed amounts for the various "recipes." Different types of scrap are used to meet the chemical requirements for different product grades and to conserve the alloy elements in the scrap. The scrap in the buckets is transferred to the furnace as part of the charge (Fig. 3).

Fluxes and Alloys

Flux materials, such as lime and fluorspar, and the alloys that are used in large quantity—such as aluminum, ferromanganese, and ferrosilicon—are stored in bulk in appropriate storage bins. Withdrawal from the bin takes place on the charging floor of the melt shop. Flux and alloy materials are drawn into charging buckets, weighed, and discharged into the furnaces.

Steelmaking Furnaces

Most of today's steel is made by the basic oxygen furnace (BOF), open-hearth furnace, and the electric-furnace processes. In the future there may be greater use of spray steelmaking and perhaps of direct reduction processes, both of which may bypass the conventional melt shop. However, the conventional melt shop will not soon be replaced. Thus the search for techniques to improve present melt-shop processes is justified.

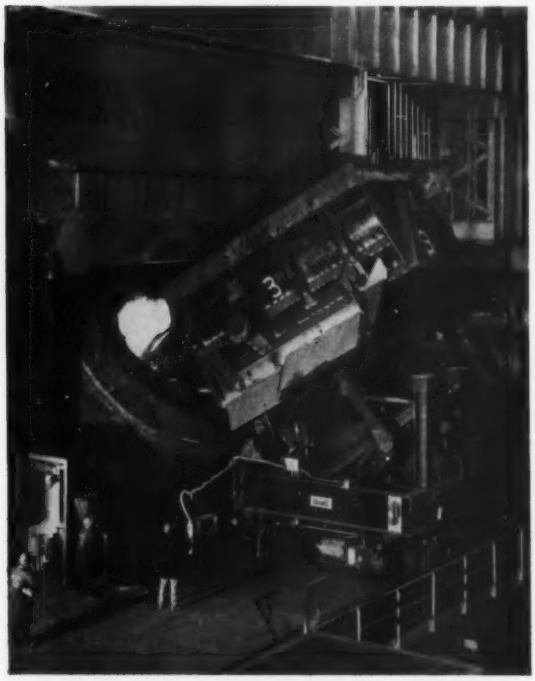

Fig. 3 Steel scrap slides from tilted charging box as first step in charging BOF.

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The BOF Process. In the BOF process (Fig. 4), high-purity oxygen is blown through a water-cooled "lance" onto liquid iron to oxidize carbon and silicon in the iron. The principal raw material is liquid blast-furnace iron, and only a limited amount of scrap can be melted since scrap is usually low in carbon. The conversion from iron and scrap into steel takes 20 to 30 min. Different configurations of the furnace are called the BOF, the Kaldo, and the Rotor furnace.

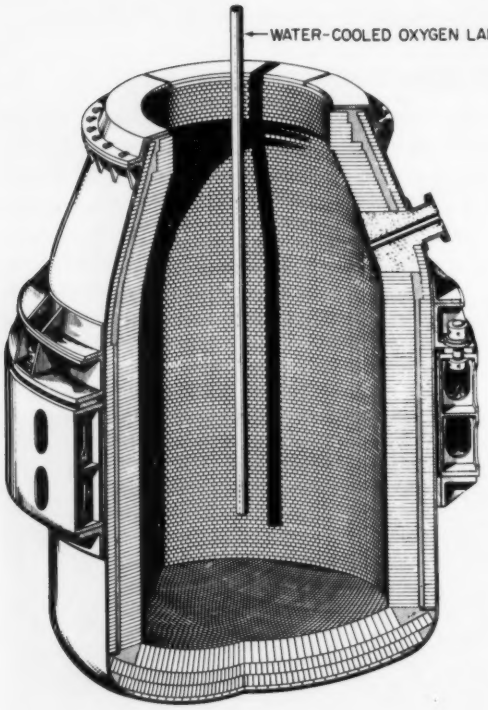

Fig. 4 Operation of the refractory-lined BOF. Pure oxygen is blown in from the top through water-cooled lances. Oxygen combines with carbon and other unwanted elements and starts a high-temperature, churning reaction that burns out the impurities from the molten iron and converts it to steel.

The Open-Hearth Furnace. The open-hearth furnace process (Fig. 5) is rapidly being supplanted by the BOF process, although it is still widely used. In the open-hearth process, the iron and scrap are melted with radiant heat from fuel. The charge may be all liquid blast-furnace iron, all scrap, or any combination; the fuel may be oil or gas. High temperatures are achieved through preheating the combustion air in brick checkerwork regenerative heaters. The furnace has two

Fig. 5 The open-hearth furnace, so named because the charge lies on a hearth where it is exposed to the sweep of flames over the surface.

such heaters, and the heat of the exhaust gas is conserved and transferred to the combustion air by periodically alternating the flow direction through preheater, furnace, and regenerator. The open-hearth process is much slower than the BOF, averaging about 11 hr. However, modern open-hearth furnaces blow oxygen on the molten steel similarly to the BOF, which halves the process time.

The Electric Furnace. For small-sized "heats," an induction-type electric furnace is often used. For heats of about 20 tons or larger, the arc-type electric furnace (Fig. 6) is used, in which heat is applied through graphite electrodes supplied with three-phase alternating current. As in the open hearth, melting in the electric furnace is done by heat; so the charge may be all scrap or any combination of liquid blast-furnace iron and scrap. Since electric energy is usually more expensive than fuel energy, the electric furnace is usually employed where its extra advantage of high temperature is required.

Most stainless and other high-alloy steels are produced by electric melting. The process typically requires 4 to 8 hr, depending on the electric power available and the amount of refining required.

Vacuum Refining

Vacuum refining (Fig. 7) reduces the concentrations of certain contaminants in steel—e.g., oxygen, nitrogen, hydrogen, carbon, and alkali metals—to

Fig. 6 Two 200-ton-capacity arc-type electric furnaces.

ultralow levels to produce a very uniform crystal structure in the steel. Ordinarily vacuum refining is a separate step after the steel is made in a furnace and may be done in the ladle or applied to a stream of liquid steel in one of several ways. If refined in a ladle, the steel must be thoroughly stirred because the vacuum is effective only at the steel-to-vacuum interface.

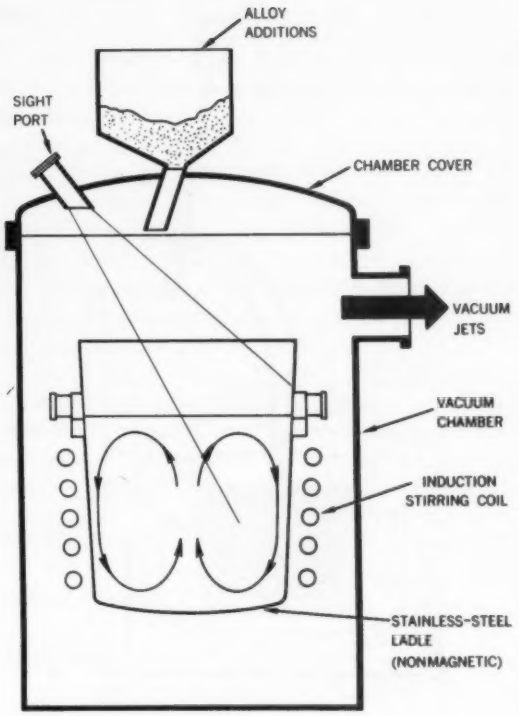

Fig. 7 Schematic of an induction-stirred vacuum-ladle refiner.

Ingot Casting

The traditional way to solidify the steel is to teem it into ingot molds (Fig. 8). Great care must be taken to have clean molds, to avoid splash, and to avoid collapsing the already solidified skin and trapping it as a foreign inclusion in the ingot. Various methods of "hot topping" are used to maintain a liquid pool at the top for the escape of gas bubbles and to fill in voids that form because of the contraction of the cooling metal. For a sound ingot structure, the ingot is usually made large and thick. The subsequent hot-rolling operations squeeze out voids and heal most cracks.

Pressure Casting

The expense and time consumed in primary rolling can be reduced by solidifying the steel directly into slabs, billets, or more complex shapes using the pressure-casting process (Fig. 9). Graphite molds, with less heat-absorbing capacity than cast-iron ingot molds, are used. The steel is forced up through the bottom of the mold into the mold cavity under pressure to obtain a sounder cast with fewer voids or cracks.

Fig. 8 Ingot casting.

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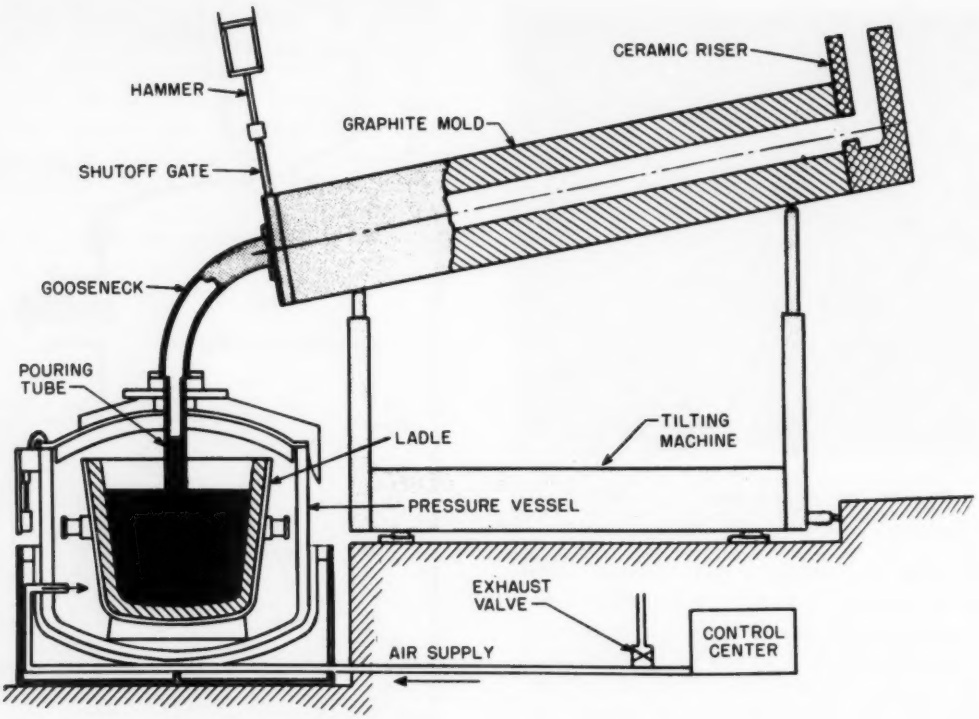

Fig. 9 Principle of operation of the bottom-pressure casting method as applied to slabs. (Courtesy of United States Steel.)

Strand Casting

Another process that bypasses the primary mill and solidifies the steel directly into slabs and blooms is the strand-casting or continuous-casting process (Fig. 10). The refined steel is teemed into an open-ended water-cooled mold where it begins to form a solid outer skin. The strand is continuously withdrawn from the bottom while liquid steel pours into the top. Below the mold, cooling water sprays are played on the strand until it has solidified all the way through. It may pass through a roll stand or two to reduce the dimensions and is then cut into convenient lengths for the next process.

ISOTOPE APPLICATIONS TO MELT-SHOP RAW MATERIALS

Radioisotopes have been used in melt-shop supply operations principally to control the uniformity of the quality and chemistry of the raw materials.

Iron

Iron is withdrawn from the blast furnace in casts or batches of several hundred tons, and its chemical composition varies from cast to cast for various reasons. Mixers are used to minimize the variation by mixing and storing a thousand tons or so of liquid iron. Radioactive tracers such as ^{59}Fe and ^{198}Au have been used to study the homogeneity achieved by the mixers. Mori and Matsuo[5] found a wide variability in mixing performance between two mixers. Williams and Howell[6] showed that a properly designed 1200-ton mixer achieves uniform dispersion of tracer in less than 20 min.

Bottle-Car and Ladle Refractory Wear

An early application of tracers was to monitor wear in iron-ladle and bottle-car linings[3] that erode from the washing action of the iron. If the liquid iron contacts the steel shell where the linings are worn through, a catastrophic spill of liquid iron could result. This is

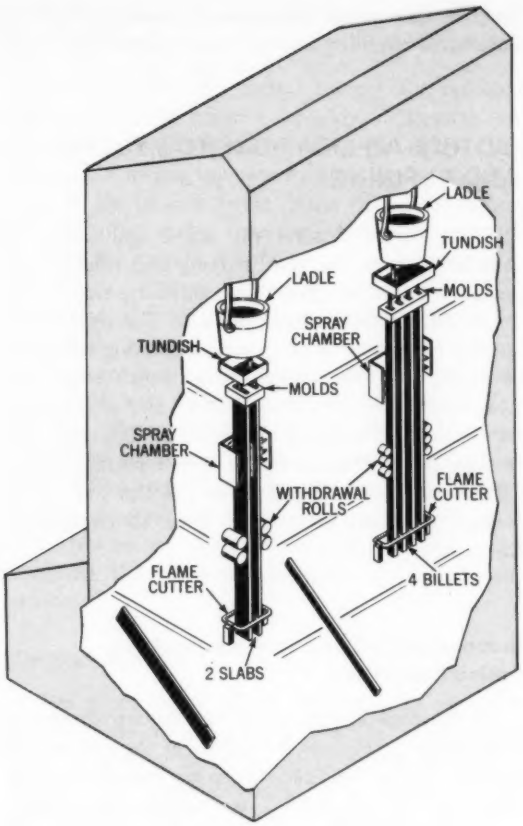

Fig. 10 Continuous-casting process.

usually prevented by constant vigilance and replacement of linings before they become dangerously worn. However, inspecting very much of a hot bottle car is difficult, and the visual inspection is subjective. The inspection needs to be made quantitative so that various types of refractories and practices can be compared. Cobalt-60 tracers have been used in various ways to monitor this wear. One way is to bury small radioisotope sources in the lining at the depth that is chosen as the maximum allowable wear for the lining. Bottle-car linings have typical wear patterns, and a few sources placed in critical spots will usually reveal the condition of the lining. The sources are small ones, too small to be detected after they dissolve in the iron; however, they are large enough to be detected from the outside when they are still in place in the lining. The lining inspector therefore looks for the disappearance of the first source.[1,7] This technique can also be applied to furnace and ladle linings.

Scrap

Most scrap is quite heterogeneous. The usual methods of chemically analyzing it depend on elaborate and tedious sampling procedures for obtaining representative samples. Through the years numerous proposals for analyzing scrap by nuclear means have been made, with the hope that a nuclear method could analyze a large sample by nondestructive and noncontact means and therefore obtain the average analysis from a representative sample. The solution to this problem is still somewhere "around the corner," but that corner seems to be coming nearer.

One project of the Republic Steel Nucleonics Division is the development of a fluorescence X-ray gage for determining the percent nickel in stainless-steel scrap. The proposed method uses a low-energy gamma-emitting radioisotope (^{109}Cd) to stimulate fluorescence X-ray emission from the scrap in a railcar or truck.[8] Once the nickel analysis has been achieved, the ability to analyze for other elements will probably be quickly added.

Ferroalloys

The use on an experimental basis of prototype equipment for fluorescence X-ray analysis in melt-shop operations has been successful. The largest electric-furnace melt shop in the industry, Republic's No. 4 Shop at Canton, Ohio, uses a tremendous quantity and variety of ferroalloy materials. Many of these look so much alike that they are sometimes incorrectly identified, especially since they are handled in bulk form without identifying labels.

An isotopic X-ray analyzer has been developed especially to classify these materials.[9] It is mounted above the conveyor belt that takes the materials from the truck hopper to the storage bins (Fig. 11). The gage

Fig. 11 An isotopic X-ray analyzer on a storage-bin conveyor.

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automatically senses the presence of material on the belt and analyzes it. This analysis is made as a considerable volume of sample runs past the gaging head. However, the analysis is completed before any material reaches the storage bin. The gage is programmed to give the operator the information that the principal element in the sample is manganese, chrome, silicon, or other. This classification is used as a check on the operator's selection of storage bin for the material. Further development will increase the sensitivity of the gage to the extent that the ferroalloy materials can be analyzed quantitatively.

Fluxes

One of the most difficult analysis problems in melt-shop practice is that of determining and controlling the basicity ratio of the slag, i.e., the ratio of calcium and magnesium to silicon in the total charge. Again, very tedious conventional methods are used to get truly representative raw-material samples and to make a very difficult chemical analysis.

Both these problems might be overcome by analyzing the flux materials with a ^{109}Cd nuclear gage above the raw-materials conveyor belt or with a similar gage on the charging floor.

There are some indications that the refractory linings of furnaces last longer when the flux materials have the correct ratio of MgO to CaO. Again, this analysis is not easy to make. The combination of tracer methods for studying refractory lining wear with nuclear noncontact methods for analyzing process materials could provide the data needed to minimize refractory costs in melt-shop operation. Another possible application for the isotopic X-ray analysis gage is to control the mixture of ingredients used in brickmaking at the refractory manufacturing stage. Gamma density gages have already been used to check the density of refractory bricks.[10]

Bin-Level Gages

Gamma-ray bin-level gaging systems are expensive and imprecise in comparison to some other types of level gages. Nevertheless, they are used widely because of their superior reliability. Many types of level gages are not suitable for service on abrasive melt-shop materials, where the gages get rough usage. Also, the type of maintenance available in a melt shop militates against complex or delicate machinery. The gamma gage works without contacting the material, and the gamma source never ceases transmitting. One particular shop in Russia uses 1216 gamma level gages[3] for an automated bin-filling and belt-feeding operation.

ISOTOPE APPLICATIONS TO MELTING AND REFINING

This section reports the major applications of radioisotopes to the steel melting and refining processes, whether in open-hearth furnaces, electric furnaces, BOF's, or vacuum refiners. The open-hearth furnace has proved to be an accommodating vehicle for a variety of radioisotope tracer experiments. The open-hearth process is very slow, and the furnace configuration allows easy extraction of samples of melt or slag. Therefore a number of very important investigations of steelmaking reaction kinetics and metallurgical processes have been done in open-hearth furnaces.

Investigations of Steelmaking Kinetics Using Radioisotope Tracers

A thorough knowledge of the reaction kinetics of steelmaking is requisite for improving the process and product quality. Radioactive tracers have been extensively used for such studies, particularly in the USSR.[3]

Radioactive ^{32}P was used to measure the kinetics of dephosphorization, ^{35}S to study the kinetics of desulfurization, and ^{59}Fe to study the kinetics of isotope exchange of iron between metal and slag. Radioisotopes of calcium (^{45}Ca) and sodium (^{24}Na) were used to investigate the phenomenon of mass transfer in slag melts. These tracers have also been used to determine the influence of stirring on the reaction kinetics, both stirring the slag only and stirring metal and slag.

Sulfur Uptake from Gas to Steel

In 1954, Russian workers reported on sulfur uptake from fuels into open-hearth steels.[1] They introduced radioactive sulfur into two types of fuel and then analyzed the metal, slag, and exhaust gas for sulfur. The fuels evaluated were fuel oil and a mixture of coke-oven and blast-furnace gases. The analyses showed that only 1% of the sulfur in the furnace atmosphere was absorbed into the metal during the refining phase but that 10 to 20% passed into the metal during melting.

Melting and Mixing Rates in Open-Hearth and Electric Furnaces

In 1955 a Russian group,[1] using ^{60}Co, reported studies of the time required for alloy additions to mix uniformly throughout the metal in an open-hearth furnace. The mixing time was different during different stages of the process, being fastest during the carbon boil. Also, the mixing time depended on the size and shape of the furnace. Therefore, open-hearth furnaces and electric furnaces have been studied with ^{51}Cr, ^{187}W, and ^{35}S to try to find optimum shapes for speeding the process.

Other experiments[1,27] with radioisotopes have determined how fast scrap melts (^{60}Co and ^{59}Fe), how fast alloys melt (^{32}P, ^{192}Ir, ^{59}Fe, ^{51}Cr, and ^{60}Co), how fast slag forms from limestone and iron ore (^{32}P, ^{140}Ba, and ^{106}Ru), and the optimum quantity of slag to retain from the prior heat (^{32}P). Also, radioactive tracer tests with radon have been used to determine the gas-flow velocity in an open-hearth furnace.

BOF Steel Analysis

The great speed of the BOF process makes it currently the lowest-cost steelmaking process; the whole cycle from tap to tap can be completed in 35 to 50 min. This speed causes many problems for the BOF operator. The 6 to 12 hr of processing time in an open-hearth furnace gave adequate time for spooning out samples, solidifying them, and sending them to the laboratory for analysis. However, the 15 to 20 min required for a complete analysis, while satisfactory for the open hearth, is much too slow for the BOF.

The current technique for rapid analysis is the use of a digital computer that is fed information through an elaborate control system (Fig. 12). All materials are analyzed before charging the BOF. This information, plus the weights of solid materials and the volume of oxygen blown, is entered into the computer, which is programmed to compute minute by minute what is happening to the charged materials and to inform the operator what actions need to be taken to produce a material with correct analysis.

Unfortunately a number of uncertainties cause errors in the calculation. One great difficulty involves getting representative analyses of scrap and flux materials; another difficulty is that of accurately determining the amount of charged carbon. Because the principal source of heat of the process is the oxidation of the carbon, the computer calculation must begin with an accurate measurement of the charged carbon.

Fig. 12 The intricate system of controls for the BOF includes a digital computer.

From gas-analysis equipment in the exhaust stack that measures the carbon in the exhaust, the amount of charged carbon that has been oxidized is calculated. This equipment is difficult to maintain, and its accuracy is low.

Every improvement in raw-materials analysis aids the BOF operator. Fluorescence X-ray analysis methods, neutron activation analysis, and inelastic-scattering methods will be used much more in the future for analyzing raw materials.

An even better possible application of radioisotopes in melt-shop applications is that of providing a noncontact instrument or method for analyzing the steel while it is in the furnace; the best possibility seems to be a neutron technique. With such an analyzer, the BOF operator would have much more precise control of the process, and the correct end result would be more certain.

Oxygen Determination in BOF Steels

Steel always picks up oxide inclusions when it is melted in vessels that are lined with oxide refractories. These oxides are detrimental to the steel because they weaken the crystal lattice and shorten the steel's fatigue life. When the oxygen process was being developed, the question was raised as to whether or not BOF steel contains more oxides than other steels. The

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measurement of oxygen content of steel in the parts-per-million (ppM) range became critical. The chemical methods first employed were laborious and gave inconsistent results.

This problem has been partially solved by activation analysis with fast (14 MeV) neutrons produced by an accelerator. The neutron technique is a much more rapid method for determining ppM amounts of oxygen in steel (less than 1 min per sample) and acts on a large volume of the sample, thus giving a more representative analysis without tedious sampling procedures.[11] The success of the neutron activation technique in the laboratory presupposes that neutron activation and inelastic scattering will be applied to the production line.[12]

Measurement of Circulation in an R–H Vacuum Degasser

Vacuum refining is effective only where the ferrostatic pressure is low. Hence some way of stirring the steel must be used to get every particle of the steel to the surface exposed to the vacuum. The Ruhrstahl–Heraeus (R–H) vacuum degasser uses argon-gas bubbles to circulate steel up through one of the two legs of a vacuum chamber (Fig. 13). The steel returns to the ladle through the other leg. Matsui et al.[13] used ^{198}Au to measure the rate of circulation in an R–H degasser. Their tests on a 100-ton ladle showed the average circulation rate to be 20 tons/min, so that all parts of the molten steel in the ladle had completely circulated in 5 min.

Measurement of Induction Stirring in a Ladle

Induction stirring is one method used to stir steel in a ladle during degassing and subsequent mixing of alloying elements. In 1965 Tucker and Radcliffe[14] developed a radioactive tracer method that did not require sampling and should therefore have other applications where sampling is difficult. They tested induction-stirring machinery for ladles of 90-, 150-,

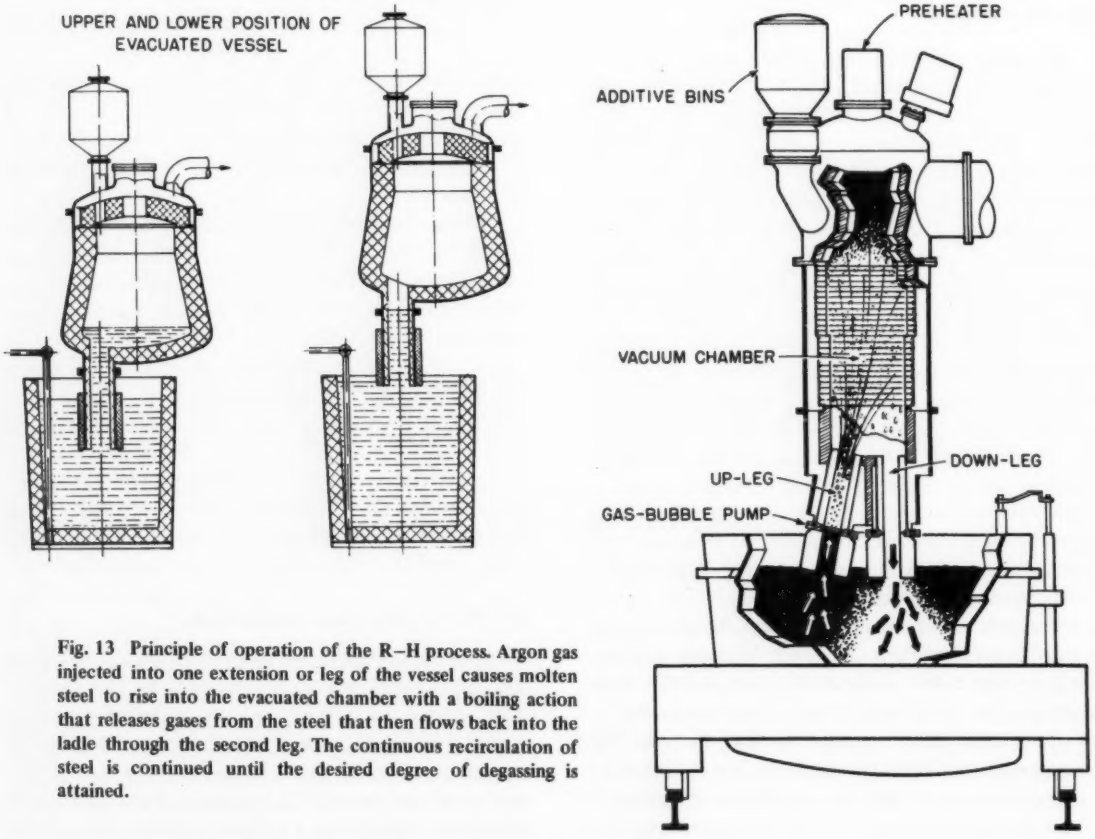

Fig. 13 Principle of operation of the R–H process. Argon gas injected into one extension or leg of the vessel causes molten steel to rise into the evacuated chamber with a boiling action that releases gases from the steel that then flows back into the ladle through the second leg. The continuous recirculation of steel is continued until the desired degree of degassing is attained.

and 200-ton capacities. The objective of these tests was to learn what configuration of coils to use and how to use the electrical parameters of current, coil turns, coil phasing, and frequency to maximize stirring effectiveness. Tracers were completely mixed in 1 to 2 min. The tracer charge used in these tests was a relatively large amount of ^{56}Mn, usually 20 to 50 Ci. The 2.11-MeV gamma rays from this tracer easily penetrate liquid steel, refractory ladle lining, and ladle shell. The tracer was put into the melt at one point, and then the time needed for the stirring system to uniformly disperse the tracer throughout the melt was determined (Fig. 14). Dispersal was determined by several detectors placed around the ladle.

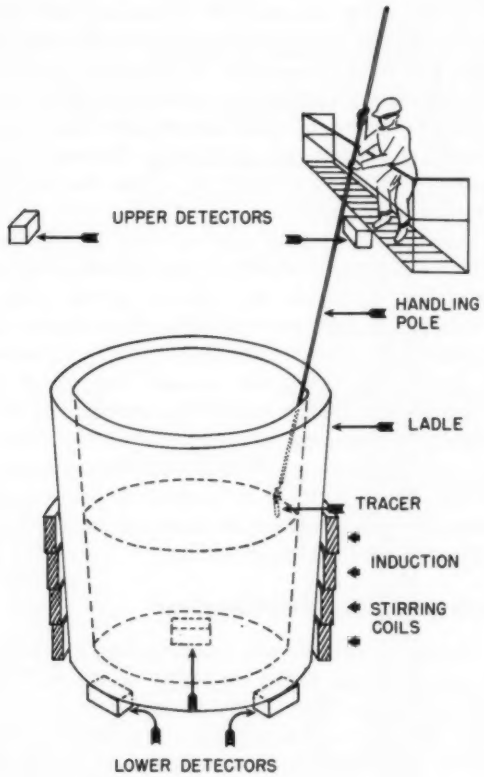

Fig. 14 Radioactive tracer tests to measure the effectiveness of induction-stirring equipment and provide factual data for future installations.

The very short half-life (2.6 hr) minimized the risk in using the very large quantity of tracer. In fact, the test program was accomplished without interference with normal operations.

Measurement of BOF Lance Position

Studies[15] on small models of the BOF, in which air was blown onto water to delineate the induced liquid flow patterns, showed that the vessel shape and the gap distance between lance tip and liquid level were critical factors. If the gap is too short, the liquid splashes out of the vessel, which results in loss of metal and fouling of the lance and hood. If the gap is too long, the stirring is poor and the result is poor utilization of the oxygen.

The liquid level varies from heat to heat because of both varying charge weight and wearing of the refractories. If the operator had good visibility, he could easily adjust the lance gap to an optimum position, but since he has poor visibility, he cannot. Some instrument must be used to determine the lance gap or the liquid level. Mechanical positioning probes have been used but are costly and inconvenient. This is a promising possible application for gamma-beam devices or gamma-ranging devices. There is also a need to corroborate in a real vessel the conclusions from the model studies. Although the model led to a good approximation of the most desirable vessel shape and nozzle arrangement, the real vessel contains a chemically reacting system with a slag layer over the steel, and the fluids have densities different from those of the modeling fluids. The nonsampling tracer technique[14] described previously could be applied to this study.

Measurement of Electric-Furnace Bottom Stirring

Because electric furnaces attain very high temperatures, they are used with alloy steels and high-melting-point steels. Elements like molybdenum and tungsten are alloyed into electric-furnace steels by adding solid lumps of alloy that slowly dissolve in the melt. In some furnaces, melt circulation is poor on the bottom of the furnace, and the molybdenum and tungsten do not fully dissolve before the heat is tapped. Faster circulation improves chemical homogeneity and alloy utilization and helps the operators determine the temperature more precisely. Therefore some electric furnaces are constructed with induction-stirring coils under the bottom for increasing the circulation. Again, the tracer test method[14] described previously could be used for studying induction stirring.

ISOTOPE APPLICATIONS TO CASTING

Casting is an entirely different procedure from melting or refining, done with different equipment by

XUM

different people. It occurs in the same shop only because once the steel is tapped from the furnace, speed is essential. There are many isotope applications in pouring and casting operations, in both ingot casting and strand casting.

Measurement of Ladle Convection Movement

The convection movement in a ladle was studied by Pocze and Folkl,[16] who used ^{67}Cu as the tracer. They tested the ladle under atmospheric pressure and also under vacuum. Kohn and Arnoult[17] reported putting some ^{198}Au tracer in a ladle a few minutes after tapping to see whether it would become uniformly mixed. Previous experiments[2] had already shown that a tracer which is unevenly distributed in the furnace becomes uniformly mixed in the ladle. Kohn and Arnoult took samples periodically during teeming. The first sample was low in gold activity, but thereafter the samples had approximately equal activity, which demonstrated that there are liquid currents in the ladle which stir the additives (something, incidentally, that steelmakers never doubted). However, this leaves the question open as to whether they arise because of convection, teeming, or the tapping momentum.

Effect of Types of Refractories and Pouring Practice

Mention has been made of determining the amount of oxide inclusions in steel by activation analysis. The presence of these oxides has led to questions about their origin, and many tracer tests and activation-analysis studies have attempted to answer them.

Calcium-45 tracers were used by Samarin[1] to determine that almost none of the inclusions originate in the slag; he found that the ladle brick is one of the major sources. Of the refractories tested, high-alumina brick caused the least contamination. Tracer tests[18] using ^{45}Ca, ^{95}Zr, and ^{59}Fe have established how much contamination comes from stopper sleeves, stopper heads, launders, nozzles, hot-top refractories, hot-top exothermic materials, etc. All these results vary, depending on the refractory used and upon the practice followed. However, an inescapable conclusion is that the greatest care must be taken with ladle and mold preparation if the cleanest possible steel is to result. Radioisotope tracers are a standard tool used to determine the sources of inclusions due to refractories.

Location of Inclusions in Cast Steel

Autoradiograms have been made from sectioned ingots labeled with ^{32}P, ^{35}S, ^{45}Ca, and ^{65}Zn to determine the distribution of inclusions and various elements of interest after solidification.[18] Other tests have determined the distribution effects of such factors as bottom vs. top pouring, mold agitation, thermal treatment to equalize composition, and rimming vs. killing.

Ingot Solidification Rate

Several authors have reported studies of ingot solidification rate using radioisotope tracers. For example, Kohn and Arnoult[17] reported introducing ^{198}Au and ^{192}Ir into the mold at various times after teeming. Results were obtained by slicing the solidified ingots and autoradiographing or by making spot-to-spot scintillation counts. The radiographs show very clearly the extent of solidification after each time interval. Large ingots took much longer than small ones to solidify. The results help to explain the general heterogeneity of ingots.

Houseman[18] used a similar method but also compared the ingot circulation currents in rimmed vs. killed steels. Rimmed steel showed greater metal movement, and there were indications that, whereas in a rimmed-steel ingot the circulation goes up the sides and down the center, the opposite happens in a killed-steel ingot. He also learned that movement of the ingots from the casting pit before complete solidification could result in an unsatisfactory microstructure, although the application of a rapid, low-amplitude vibration during solidification improved mixing and slightly shortened solidification times.

Liquid Level in the Strand-Caster Mold

During the next decade, strand casting will probably replace ingot casting as widely as the BOF is replacing the open hearth. The strand caster complements the BOF very well because the BOF delivers heats on a regular schedule—one heat every 45 min or so—and the strand caster can be matched to this schedule. In an electric furnace or open-hearth melt shop, the heats tend to tap in random fashion so that the furnaces at times have to hold back for the strand caster while at other times there is no steel ready to cast.

One objective in strand-casting control is the maintenance of a constant level of liquid steel in the mold. Gamma-ray level sensors are widely used for this purpose. One of their advantages over other types of level sensors is that they can be separate from the mold and thus measure absolute liquid elevation rather than relative level in the oscillating mold.

Extent of Liquid Center in Strand Castings

Because steel is a relatively poor heat conductor, a liquid center persists in the strand below the mold. Cutting to length must be done at a point below the end of the liquid center. Thus the need exists to determine how far this center extends for various strand dimensions and casting speeds. Varga and Fodor[19] did this by dropping a tungsten-encased ^{60}Co source into the mold to see how far it would sink into the liquid pool (Fig. 15). Another method, used by Polish workers,[20] was to add radioactive phosphorus or cobalt to the molten pool and later section and autoradiograph the strand. The autoradiographs revealed the average skin thicknesses at various levels and speeds of casting.

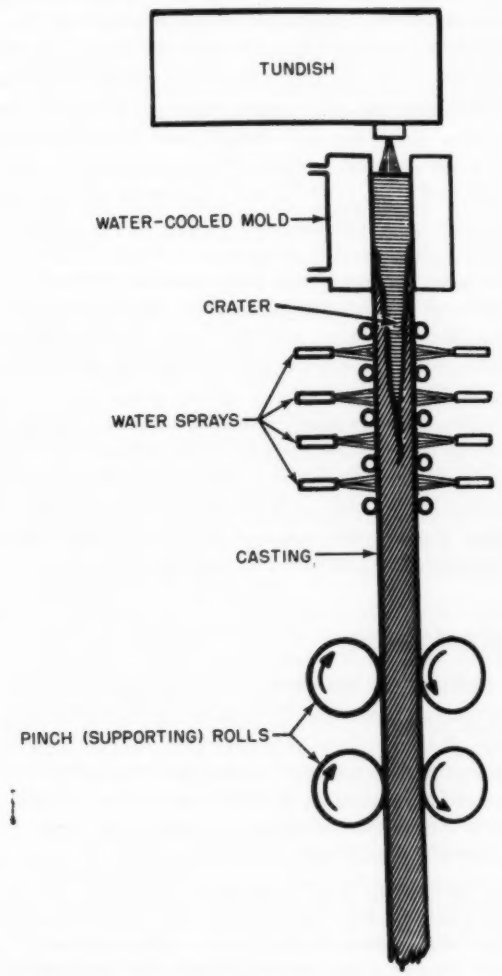

Fig. 15 Schematic diagram depicting how liquid metal persists in the center of a continuous casting for a considerable distance after the casting leaves the water-cooled mold, forming a so-called "crater." (Courtesy of United States Steel.)

MISCELLANEOUS MELT-SHOP APPLICATIONS

Additional applications of radioisotopes in melt-shop operations are varied.

Quick Chemical Analysis in BOF Shops

The critical need for quick chemical analyses was pointed out earlier. The furnace itself is a poor environment for any instrument, and an ideal in-the-furnace analyzer may be impractical. Metallurgists struggle along with analyses done on samples.

Many fluorescence X-ray machines are being used in melt shops for steel analysis (Fig. 16). An X-ray tube

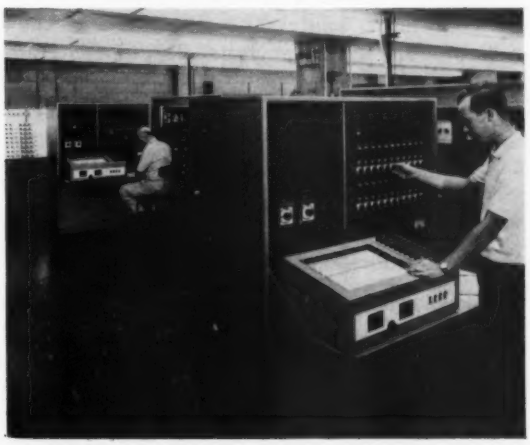

Fig. 16 Control laboratory for rapid spectrographic analysis of steel samples.

produces the exciting X rays, which have a somewhat continuous spectrum up to a certain maximum energy determined by the accelerating voltage used in the tube. The fluorescence X rays coming from the samples are collimated into a narrow beam and dispersed from a crystal into the different energy levels or spectral bands. The yield in counts detected is very small because the continuous spectrum does not excite the characteristic X rays of the sample very efficiently, and the collimating and crystal diffracting system can only

XUM

use a very minute fraction of the fluorescence X rays that the sample generates. Patent applications[21] and published papers[22] suggest that isotopes may replace X-ray tubes in spectrographs with a consequent improvement in speed and accuracy of analysis. Furthermore, gamma emitters generate a line spectrum that is much better than a continuous spectrum for some purposes. For example, if the exciting gamma energy falls between the characteristic X-ray energies of two constituents, the higher-energy element will not be excited. As a result, a small amount of lower-energy element in a matrix of a higher-energy element can be accurately measured, e.g., chromium in an iron matrix.

In some cases there are no known isotopes that emit gammas of the ideal energy for exciting the element of interest. However, with proper target elements and filters, bremsstrahlung sources can be made that emit most of their gammas in a fairly narrow energy band.[23]

Another trend is toward nondispersive techniques of fluorescence X-ray detection. The X rays of all energies are detected with the same detector, and the energy sorting is done electronically instead of with a collimator and dispersion crystal. This technique uses the fluorescence X rays much more efficiently; so its use should speed up X-ray analysis. Since a typical steel-plant chemistry laboratory may do 1200 or more analyses per day, a few seconds saved per sample is very important.

Material Identification

Lindblom, Erwall, and Murray[24] reported the use of radioactive tracer additions to provide positive identification of materials destined for critical use in nuclear reactor operations. The objective of the "fingerprinting" was to make sure that the specially prepared heats were used as intended and not inadvertently mixed up in subsequent production operations.

Tantalum-182 was added to the stainless-steel melts before pouring. The individual ingots were then followed through production operation by means of radiation detectors. Similarly, when the ultimate tubular product was to be shipped to the reactor for installation, each tube could be identified as belonging to the shipment.

The technique provides a positive method for identifying a special heat. Indeed, by using several tracers in various combinations, a number of different heats could be identified. However, this technique may not be practical for the ultimate application—i.e., identifying all steel heats in a mill. The principal difficulties are: (1) the large number of heats and the large number of different steel types, (2) the relatively small selection of isotopes having the correct energy and half-life, and (3) the buildup of residual background of some tracers caused by recycling of the scrap. The long-range solution to this need would seem to be an inexpensive, simple analyzer of the X-ray or neutron type for use at routing and controlling points in the steel plant.

Weighing by Radioisotope Dilution

Ten years ago when the open-hearth process was the predominant steel process, alloying was done in increments. The weight of the melt did not have to be known beforehand because it was determined from the dilution of the first alloy-addition increment. When the high-volume use of oxygen began to speed up the process, there was not enough time for incremental alloying. The melt weight had to be known accurately. Therefore ^{65}Ni ($T_{1/2}$ = 2.56 hr) was proposed for routine use for determining the weight of the steel in a furnace through the principle of tracer dilution.[25] However, not many melt shops were close enough to a source of radioisotope tracers to be able to use such a short-half-life tracer, and the rapid improvement of strain-gage load cells has since obviated the need to use tracer dilution.

The determination of the amount of slag in a furnace is very difficult to do by any means other than tracer dilution. Erwall and Ljunggren[26] reported that lanthanum has been used for measuring slag weight as either a radioactive tracer or an inactive tracer, with the sample being activated for analysis later.

CONCLUSION

This article has covered the application of radioisotopes and radiation technology to the whole melt-shop domain—from raw materials, through furnace operations, to pouring and casting operations—and has included product quality controls and chemical tests. Numerous radioisotope applications that are incidental to melt-shop operations will be covered in subsequent articles (emergency markers, crane anticollision devices, and dust-emission tracing). While much has already been accomplished with radioisotopes, there remains a vast potential for further applications. A cautious projection of a few future developments has been given. This glimpse of the past and the future of radioisotope applications in modern steelmaking melt shops hopefully will stimulate increased management

support for developing the men, facilities, and programs to accomplish the potential of radioisotope utilization. (FEM)

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

1. A. M. Samarin, The Use of Artificially Radioactive Isotopes in the Study of the Processes of the Production of Steel and Iron, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955*, Vol. 15, pp. 151-159, United Nations, New York, 1956.
2. N. G. Bogdanova, P. L. Gruzin, G. I. Yermolayev, and I. D. Nikulinsky, The Use of Radioactive Isotopes in Metallurgical Research, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 19, pp. 180-186, United Nations, New York, 1958.
3. P. L. Gruzin, Yu. F. Babikova, G. S. Gerasimchuk, A. K. Lebedev, G. S. Rozhavskii, and G. B. Federov, Current State and Trends in the Use of Radioisotopes and Nuclear Radiations in Metallurgy and Mining, USAEC Report AEC-tr-6466/3, pp. 111-120, 1966; a translation of The Uses of Radioactive Isotopes and Nuclear Radiation in the U.S.S.R., Proceedings of the All-Union Conference, Riga, 1960, Vol. III.
4. E. Booth, Radioisotopes in the Steel Industry, *At. Energy Aust.*, **11**(3): 9-12 (July 1968).
5. *H. Mori and M. Matsuo, Study on Homogeneity of Pig Iron in Mixer—Radioisotope Tracer Technique, *Yawata Tech. Rep.*, No. 247: 5025-5028 (June 1964).
6. K. F. Williams and E. D. Howell, The Use of Radioisotopes to Determine Molten Metal Mixer Efficiency in Steelmaking Practice, in *Radioisotope Tracers in Industry and Geophysics*, Symposium Proceedings, Prague, 1966, pp. 641-650, International Atomic Energy Agency, Vienna, 1967 (STI/PUB/142).
7. Y. Lindblom and N. Stahl, Wear Measurements on the Lining of a Kaldo Converter (in Swedish), *Jernkontorets Ann.*, **150**: 323-327 (1966); also in L. G. Erwall, Use of Radioisotope Techniques in Sweden, *Isotop. Radiat. Technol.*, **4**(3): 255-258 (Spring 1967).
8. *S. J. Aron, X-Ray Fluorescence Analysis of Iron–Nickel Binary Using 60 keV Gamma Radiation, Republic Steel Corporation Report ERC-1799, June 1969, unpublished.
9. *W. R. Pavlik, Ferroalloy Gage at Canton #4 Melt Shop—Conclusion of Prototype Development Program, Republic Steel Corporation Report ERC-1766, November 1968, unpublished.
10. J. J. Borbas, R. C. Padfield, and E. Moscker, Density Determination of Refractories by Measurement of Gamma-Radiation Absorption, *J. Amer. Ceram. Soc.*, **50**(8): 421-424 (Aug. 21, 1967).
11. A. L. Gray and A. Metcalf, Industrial Applications of Neutron Activation, in *Proceeding of the 1965 International Conference on Modern Trends in Activation Analysis*, pp. 86-90, College Station, Texas, 1965, Texas A&M University, College Station, 1965.
12. J. R. Rhodes, Applications of Neutron Activation to On-Stream Analysis, *Isotop. Radiat. Technol.*, **6**(4): 359-368 (Summer 1969).
13. *K. Matsui, M. Nakayama, R. Arima, N. Yamazaki, and K. Asano, R–H Circulating Flow Degassing Process, *Tech. Rep. Fuji Iron & Steel Co., Ltd.*, **13**(4): 597-614 (December 1964); also published as Technical Report, Overseas No. 1, Fuji Iron & Steel Co., Ltd., May 1965.
14. W. B. Tucker and T. J. Radcliffe, Use of Nuclear Tracers for Measurement of Induction Stirring Dynamics in Vacuum Degassing Operations, prepared for presentation at the AISE Technical Program, September 1970, Iron and Steel Exposition, Cleveland, Ohio.
15. *T. Simotsuma and K. Sano, Effect of Bath Motion on Splashing of Basic Oxygen Furnace Bath, Part II. A Study of Various Phenomena in Basic Oxygen LD Furnaces, Based on Room-Temperature Models (in Japanese), *Tetsu To Hagane*, **51**(10): 1909-1911 (1965).
16. L. Pocze and O. Folkl, Ladle Behavior of Molten Steel Under Atmospheric Pressure and Vacuum Using Radioactive Isotopes (in Hungarian), *Kohasz. Lapok*, **97**(3): 153-155 (April 1964).
17. A. Kohn and J. Arnoult, Examination of Convection Currents of Killed Steel During Solidification (in French), *Rev. Met. (Paris)*, **62**(4): 311-320 (April 1965).
18. D. H. Houseman, Radioisotope Studies of Steel Ingot Solidification, *Steel Times*, **192**(5106): 689-695 (May 27, 1966).
19. C. Varga and J. Fodor, The Use of Radioactive Isotopes to Inspect the Technology of the Continuous Casting Process, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 19, pp. 235-236, United Nations, New York, 1958.
20. *H. Firganek, J. Labus, B. Paczula, J. Siewierski, and K. Raczka, Investigation of Solidification Front and the Depth of the Liquid Phase in Continuously Cast Billets by the Technique of Radioactive Tracers (in Polish), *Prace Inst. y Tutow Hutniczych*, **19**(6): 373-379 (1967).
21. J. S. Watt, Method and Apparatus for Producing X-Rays of Particular Wave Lengths and Applications Therefor, Canadian Patent No. 758,195 (to Australian Atomic Energy Commission), May 2, 1967.
22. M. Gerrard, Potential Markets for Radioisotope-Based Analytical Equipment, *Isotop. Radiat. Technol.*, **4**(3): 366-369 (Summer 1967).
23. J. J. Ezop and T. G. Stinchcomb, Study, Design and Applications of Beta-Excited X-Ray Sources, in *Proceedings of the Symposium on Low-Energy X- and Gamma Sources and Applications*, Illinois Institute of Technology Research Institute, Chicago, 1964, USAEC Report ORNL-IIC-5, pp. 4-37, November 1965.
24. Y. Lindblom, L. Erwall, and F. Murray, Labelling of Metallic Materials with Radioactive Tracer Elements (in Swedish with English abstract), *Jernkontorets Ann.*, **150**: 277-297 (1966); also in L. G. Erwall, Use of Radioisotope Techniques in Sweden, *Isotop. Radiat. Technol.*, **4**(3): 255-258 (Spring 1967).
25. *R. A. Schmitt and R. A. Sharp, Use of Short-Lived Radioisotope 2.56-Hr ^{65}Ni in Determining the Melt

XUM

Content in Open-Hearth Furnaces, Report GA-1083, General Atomic Division, General Dynamics Corp., June, 1960.

26. L. G. Erwall and K. Ljunggren, Recent Uses of Radioisotopes in Technological Research and Control in Sweden, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 19, pp. 3-12, United Nations, New York, 1958.
27. Yu. S. Paniotov and Yu. N. Yakovlev, Study of Smelting and Slag Formation in Large Loaded Open-Hearth Furnace by Means of Radioisotopes (in Russian), in *Metallurgiya i Koksokhimiya, Sbornik Statei*, No. 7, pp. 59-64, Publishing House "Tekhnika," Kiev, 1967.

Inspection of Hand-Grenade Fuses with a Radioactive Isotope*

Summarized by Martha Gerrard

Abstract: *A method for inspecting the delay-column height (M200 series) in hand-grenade fuses was developed in which a tracer quantity of a radioactive element is incorporated in the delay composition. The gamma radiation from each fuse is measured to give data from which the total delay-charge weight is calculated. Tests on the method with plant-size batches of delay composition indicated that an inspection rate of two units per second can be achieved without exceeding permissible external radiation dose levels.*

The proper length, or total weight, of the delay-charge element in a hand-grenade fuse is critical. Normally the charge is loaded into the delay housing (Fig. 1) by a series of incremental fillings and compactions, performed either manually or automatically. To determine whether or not the housing is properly filled, the density of the charge is determined by a technique in which a fraction of the delay-column length is viewed by a collimated beam of X rays. The transmitted radiation, a function of the charge density, is determined by a cadmium sulfide detector. However, this method does not provide the necessary degree of quality control because the entire delay column is not viewed, and a fuse may pass inspection even though part of the delay charge is missing.

If a radioactive tracer is incorporated uniformly in the delay composition, the intensity of the gamma-ray signal from each delay column should be a measure of the total charge weight and hence of the completeness of filling of the column. The radioactive tracer element would not have to be common to the delay-composition ingredients but could be added as a foreign element in the ppM concentration. Thus radioactive elements of a wide range of radiation half-lives and gamma-ray energies could be used. A study of the feasibility of inspecting M200 fuses by such a technique was therefore undertaken. The results of some tests on the technique are reported here.

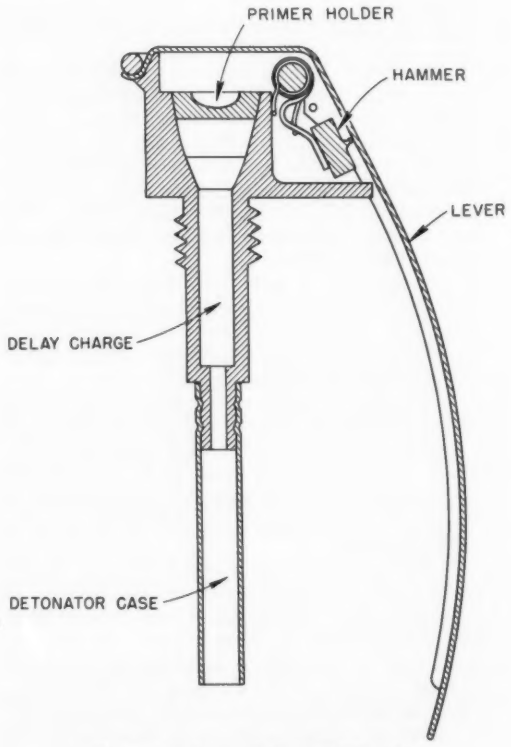

Fig. 1 Hand-grenade fuse (M200 series).

*Summary of "A Radioactive Tracer Method for the Inspection of the Delay Element in M200 Series Hand-Grenade Fuzes" by Stanley Semel, Martin Gilford, and Samuel Helf, Feltman Research Laboratories, Picatinny Arsenal, Dover, N. J., Technical Report 3911, September 1969.

RADIOACTIVITY REQUIRED FOR A GIVEN INSPECTION RATE

The amount of radioactive tracer, R (μCi), required per fuse for an inspection rate of one unit per second, with the requirement that the charge weight be determined to ±5% accuracy, is found from

$$R = 800/GFASK$$

where G = number of gamma rays emitted per disintegration
F = efficiency of detector for sensing gamma rays
A = gamma attenuation factor of fuse-body housing
S = gamma self-shielding factor of delay column
K = disintegration constant = 3.7×10^4 per sec per μCi

The value for G is obtained from the decay scheme of the radioactive element selected, and F is determined by experiment with known gamma emitters for a particular detector. The value of A is calculated from $A = e^{-\mu\rho t}$, where μ is the mass absorption coefficient of the delay-column housing in cm^2/g, ρ is the density of the housing, and t is the thickness through which the radiation passes. The appropriate value for μ is obtained from the composition of the absorbing material and the specific gamma-ray energy. The value of S can be estimated, by the method of Rockwell,[1] from the composition of the delay powder, its physical geometry, and the gamma-ray energy.

The equation is derived as follows: The standard deviation in N total counts is $\pm\sqrt{N}$ and the percent error $= \pm 100\sqrt{N}$. For a 5% error, $\sqrt{N} = 20$ and $N = 400$. Therefore 400 counts are required from each radioactive fuse. Assuming that, in a 1-sec interval, half the time will be required for counting and half for decision making (pass or fail), a count rate of 800 counts/sec would be needed.

In an automatic inspection process with radioactive delay composition, the pass or fail criterion will depend on a measured difference of 20% in count rate. If the inspection safety factor is doubled, this pass or fail criterion can be set at 10% count-rate difference. Thus, for an inspection speed of one unit per second, where a fully loaded delay column gives a count rate of 800 per sec, a count rate of 720 per sec or less will cause rejection.

Two radioactive tracers—^{51}Cr and ^{134}Cs—were selected for study. For an inspection rate of one unit per second, 0.62 μCi of ^{51}Cr or 0.022 μCi of ^{134}Cs would be required (Table 1).

TEST RESULTS

For these tests, two batches of delay composition were prepared with each tracer, one a 2000-g laboratory batch and the other a 30-lb batch to simulate actual production quantities. The levels of radioactivity per fuse were about half those required for an inspection rate of one item per second, i.e., 0.3 μCi of ^{51}Cr and 0.01 μCi of ^{134}Cs.

As was expected, the count rate for loaded, labeled delay columns decreased essentially linearly as the delay-column height decreased (Fig. 2). This was true for both ^{51}Cr- and ^{134}Cs-labeled material and for both laboratory- and plant-size batches of delay charge. Thus an inspection system based on counting the total radioactivity in hand-grenade fuses filled with radioisotope-labeled delay composition would readily reject incompletely filled units.

In the experiments, type I Zr–Ni alloy delay composition—60/14/26 $BaCrO_4/KClO_4$/70-30 Zr–Ni—was used. The ^{51}Cr was added in the form of $Ba^{51}CrO_4$ as a fractional percentage of the total barium chromate, and the ^{134}Cs was included (as $^{134}CsClO_4$) in ppM concentrations in the $KClO_4$.

Each fully loaded column required 1333 mg of delay powder pressed as four 333-mg increments at

Table 1 Factors for Estimation of Amount of Tracer per Fuse for an Inspection Rate of One Unit per Second

Radioactive tracer	Half-life	E_γ, MeV	G	F*	S	A	R
^{51}Cr	27.8 days	0.32	0.09	0.5	0.92	0.85	0.62
^{134}Cs	2.1 years	0.75†	2.26	0.5	0.95	0.90	0.022

*Assuming that each fuse is counted between two 2- by 2-in. NaI(Tl) scintillation crystals.
†Average of six gamma rays.

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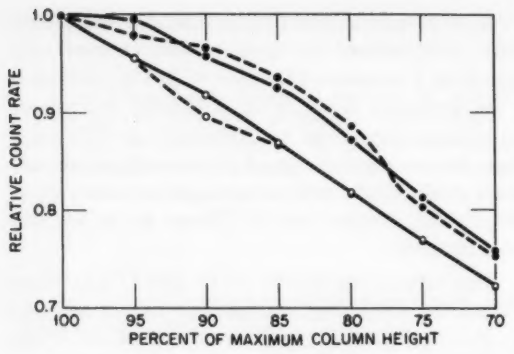

Fig. 2 Count rate as a function of column height for (○) ^{134}Cs- and (●) ^{51}Cr-labeled delay charge for hand-grenade fuses. ——, 2000-g batches; – – – –, 30-lb batches.

approximately 20,000 psi consolidating pressure. The increment weights and the number of increments were chosen to give column heights between 70 and 100% of maximum. To ensure an accurate transfer position to the fuse bodies, column heights were measured for all items loaded. One hundred elements were loaded for each of the four delay-composition batches. No detonator or igniter assemblies were connected to the fuse bodies.

Gamma radioactivity measurements were made with a NaI well-crystal integral-line scintillation counting assembly.

SAFETY CONSIDERATIONS

External radiation levels from the mixing operation of 30-lb batches of delay composition, for both ^{51}Cr- and ^{134}Cs-labeled mixes, were well within the accepted minimum permissible value of 2.5 mRems/hr for radiation workers (Table 2). These are sufficiently low that the radioactivity concentration can be safely doubled to allow for an inspection rate of two units per second. These levels are also of the same order of magnitude as obtained from the use of X-ray and nuclear inspection gages. Thus, with respect to external radiation, the radiotracer method presents no special safety problem.

Table 2 External Radiation Measurements for 30-lb Delay Compositions

Location	Radioactivity, mRem/hr	
	^{51}Cr	^{134}Cs
Against mixing vessel	0.1	1.0
12 in. from mixing vessel	Background	0.2
Over open mixing vessel, at lip	0.2	0.6
Delay composition, at surface	0.6	1.0

REFERENCE

1. Theodore Rockwell III, (Ed.), *Reactor Shielding Design Manual*, McGraw-Hill Book Company, Inc., 1956.

Use of Radioisotopes in Detector Calibrations

By J. H. McCrary*

Abstract: *A means for measuring detector sensitivities to monoenergetic electromagnetic radiations is described. The accuracy of the results should be approximately ±10%, which is comparable with measurements now being made with X-ray machine sources.*

This article describes the use of radioisotopes for calibrating detectors. Certain experiments require the use of photon detectors that are relatively insensitive but whose energy sensitivity function is well known, e.g., the photoelectric diode (XRD) and the fluor photodiode (FPD). The sensitivity of each detector must be measured over its useful energy range, which is typically between about 5 and several hundred keV. Sensitivity measurements are currently being performed using X-ray machines, fluorescers, and filters.

*EG&G, Inc. (Edgerton, Germeshausen and Grier, Inc.), Las Vegas Division, 680 E. Sunset Rd., Las Vegas, Nev. 89101. This is a revision of Technical Memorandum No. EGG 1183-1401, Aug. 27, 1968.

While these techniques produce adequate calibration of the detectors, it seems advisable to have additional calibration facilities in the form of radioactive sources. A calibration facility using radioisotopes would supplement the present measurement facility by extending the energy range to higher energies and provide both a standby facility for use when the X-ray machines are inoperable and a means for checking the accuracy of the free-air ionization chamber currently used to measure X-ray-beam energy fluxes. The calibration of detectors on a different facility would provide an independent verification of the results obtained with the present facility. Finally, the radioisotope facility would be small and reliable enough for use in the field.

SOURCE REQUIREMENTS

A photon source useful for calibrating detectors must possess one or, at most, two emission lines of gamma or X rays of useful energy. The emission spectrum must be clean and free of radiations other than those being used. The radioisotope should have a useful half-life—preferably from 1 month to 1 year—and a high specific activity and small volume to prevent scattering and attenuation within the source. The source must be intense enough to produce a measurable current in the detector.

Let us consider a 1-Ci source of 100-keV photons in a geometry where the detector subtends, at the source, a solid angle equal to 1% of a sphere. The energy flux, ϕ, reaching the detector is

$$\phi = \frac{(3.7 \times 10^{10})(1.602 \times 10^{-9})(100)(0.01)}{\text{detector area}}$$

where the first numerator term is in photons per sec, the second in ergs per keV, the third in keV per photon, and the fourth in hundredths of the solid angle. For a 1-in.-diameter detector, $\phi = 11.7$ ergs/(cm^2)(sec), which would produce a net current of about 5×10^{-14} A in an XRD or about 5×10^{-12} A in an FPD. This is a usable net current for an XRD calibration, while 10^{-12} A is sufficient for an FPD. Therefore a source of ~250 mCi would be sufficient for FPD calibration purposes. Figure 1 is a plot of required source strength vs. energy for XRD (nickel cathode) and FPD calibrations. One-percent geometry is assumed, as are net currents of 10^{-13} A for the XRD curve and 10^{-12} A for the FPD curve. For shielding of sources in the few-curie range, 1 in. of lead is sufficient for photons with energies below 200 keV. Higher-energy sources would require more elaborate shielding.

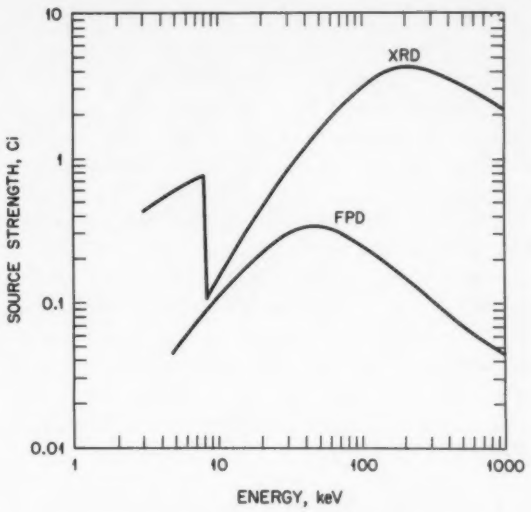

Fig. 1 Radioisotope source strength required, as a function of energy, for use in calibrating XRD (nickel cathode) and FPD detectors.

POSSIBLE SOURCES

Several possible photon sources are discussed below, arranged in order of increasing atomic number. The list is not complete, but it contains most of the promising candidates. The quoted X-ray energies are the weighted means of the K_α and K_β energies. The data are taken from Refs. 1–6.

^{51}Cr: Decays 91% by electron capture to the ground state of ^{51}V and 9% to the 320-keV first excited state of ^{51}V. This 9% efficient source of 320-keV gamma rays has a half-life of 27.8 days. Also present are 5-keV vanadium X rays, which could be either filtered out or used. Chromium-51 can be produced in a reactor from ^{50}Cr, which has a thermal capture cross section of 17 barns. Since ^{50}Cr is only 4.3% abundant, chromium enriched in this isotope should be used.

^{55}Fe: Decays 100% by electron capture to the ground state of ^{55}Mn to emit the 5.9-keV manganese X rays. The half-life of ^{55}Fe is 2.6 years. Iron-55 is produced by the ^{54}Fe$(n,\gamma)^{55}$Fe reaction in a nuclear reactor. The thermal-neutron cross section for this reaction is 2.5 barns. Iron-54 is 5.82% abundant in natural iron. Iron-58, which is 0.33% abundant, leads to spectral impurities. It is therefore advisable to irradiate iron enriched in ^{54}Fe rather than natural iron in producing this source.

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^{60}Co: Beta decays more than 99% to the 2.51-MeV state in ^{60}Ni. The deexcitation of this state results in 1.173- and 1.332-MeV gamma rays of equal intensity. The half-life of ^{60}Co is 5.26 years. Cobalt-60 is produced by irradiating natural cobalt (100% ^{59}Co) in a nuclear reactor. The thermal capture cross section is 37 barns.

^{71}Ge: Decays 100% by electron capture to the ground state of ^{71}Ga, which emits 9.4-keV gallium X rays. The half-life of ^{71}Ge is 11.4 days. It can be reactor produced by the ^{70}Ge$(n,\gamma)^{71}$Ge reaction, with a thermal cross section of 3.5 barns. Germanium-70 is 20.5% abundant. Because of this and possible problems from other germanium isotopes, it is recommended that sources be prepared from enriched ^{70}Ge.

^{109}Cd: Decays by electron capture to the 87.7-keV isomeric state in ^{109}Ag. This state decays (in 40 sec) 5% of the time by gamma-ray emission. Cadmium-109 emits 22.6-keV silver X rays with 100% efficiency and 88.1-keV gamma rays with 5% efficiency, with a half-life of 453 days.[7] The X ray could be either filtered out or used in a double calibration technique. Cadmium-109 is produced in three ways:

$$^{108}\text{Cd}(n,\gamma)^{109}\text{Cd} \text{ (reactor neutrons)}$$

$$^{109}\text{Ag}(p,n)^{109}\text{Cd} \text{ (cyclotron protons)}$$

$$^{107}\text{Ag}(n,\gamma)^{108}\text{Ag} \xrightarrow{\beta} {}^{108}\text{Cd}(n,\gamma)^{109}\text{Cd} \text{ (reactor neutrons)}$$

The first reaction appears to be the most promising. Because of spectral impurities from heavier cadmium radioisotopes, highly enriched ^{108}Cd should be used. Cost and production time are the main disadvantages of this source.

^{125}I: Decays 100% by electron capture to the 35.5-keV state in ^{125}Te. The source emits tellurium X rays (28.1 keV) with each decay and 35.5-keV gamma rays in 7% of the decays. The half-life is 60 days. Iodine-125 is the daughter of ^{125}Xe, which is reactor produced by the ^{124}Xe(n,γ) reaction.

^{137}Cs: Beta decays 93.5% to ^{137m}Ba with a half-life of 30.0 years. Barium-137m emits a 662-keV gamma ray 89% of the time in decaying to its ground state. There are no spectral contaminants other than bremsstrahlung. Cesium-137 is a ^{235}U fission product and is produced by chemical separation from spent reactor fuel elements.

^{141}Ce: Beta decays in 33 days to ^{141}Pr. Seventy percent of the beta decays go to the first excited state of ^{141}Pr, which emits a 145-keV gamma ray. The remainder of the decays go to the ground state. Cerium-141 is produced by reactor irradiation of natural CeO. The material typically contains about 1% ^{139}Ce, which emits a 166-keV gamma ray. This is not considered a serious problem.

^{198}Au: Decays by beta emission to the first excited state of ^{198}Hg at 412 keV 99% of the time. The half-life is 2.70 days. The beta spectrum end point is 960 keV. There are weak (less than 1%) lines at 1.09 and 0.68 MeV. High specific activity and relatively low-cost sources can be made by reactor irradiation of natural gold (100% ^{197}Au).

^{203}Hg: Decays 100% to the first excited state of ^{203}Tl, which emits 279-keV gamma rays 77% of the time. The beta decay end point is 213 keV, and the only spectral impurities are bremsstrahlung. The half-life of ^{203}Hg is 46.9 days. It is produced by the reactor irradiation of natural mercury. Mercuric oxide could be used to prevent problems associated with liquid sources.

^{241}Am: Decays in 458 years to ^{237}Np, which emits a 59.5-keV gamma ray 36% of the time. Other extremely weak gamma rays will not present any serious problems. Americium-241 is readily available as a by-product of spent fuel elements.

For isotopes that emit photons of two well-separated energies, simultaneous sensitivity measurements can be made at the two energies, the output being measured with and without a filter in the beam. Let

E_1 and E_2 = energies of the two lines
I_1 = current without filter
I_2 = current with filter
S_1 = sensitivity at energy E_1
S_2 = sensitivity at energy E_2
μ_1 = filter attenuation coefficient at E_1
μ_2 = filter attenuation coefficient at E_2
ϕ_1 = flux at E_1
ϕ_2 = flux at E_2
x = filter thickness

Then $I_1 = S_1\,\phi_1 + S_2\,\phi_2$

$$I_2 = S_1\,\phi_1\,e^{-\mu_1 x} + S_2\,\phi_2\,e^{-\mu_2 x}$$

Since all the above quantities are known except S_1 and S_2, the solution for the sensitivities is

$$S_1 = \frac{\begin{vmatrix} I_1 & \phi_2 \\ I_2 & \phi_2\,e^{-\mu_2 x} \end{vmatrix}}{\begin{vmatrix} \phi_1 & \phi_2 \\ \phi_1\,e^{-\mu_1 x} & \phi_2\,e^{-\mu_2 x} \end{vmatrix}}$$

and

$$S_2 = \frac{\begin{vmatrix} \phi_1 & I_1 \\ \phi_1\, e^{-\mu_1 x} & I_2 \end{vmatrix}}{\begin{vmatrix} \phi_1 & \phi_2 \\ \phi_1\, e^{-\mu_1 x} & \phi_2\, e^{-\mu_2 x} \end{vmatrix}}$$

It is important that the lines be well separated in energy and that they be of nearly the same intensity. This unfolding technique should work for any number of lines, but it is not recommended for accurate sensitivity measurements for more than two.

EXPERIMENTAL PROCEDURE

It is desirable to establish a simple standard geometry that can be reproduced in the laboratory or in the field. Materials that would scatter photons and thus distort the spectrum must be kept to a minimum. In Fig. 2 is shown a geometry where the source is contained in a thin-walled aluminum capsule, which is mounted atop a tripod that holds the source rigidly 2.5 in. above the sensitive surface of the detector. Air scattering can be minimized by attaching one or two lead collimators to the tripod legs. Filters, when needed, can be placed over the face of the detector. The proximity of the source to the detector causes the detector to see a divergent photon beam with a half angle of 11.3°. It is believed that the only consequence of this divergence is an effective slight (2%) thickening of the detector window at its periphery.

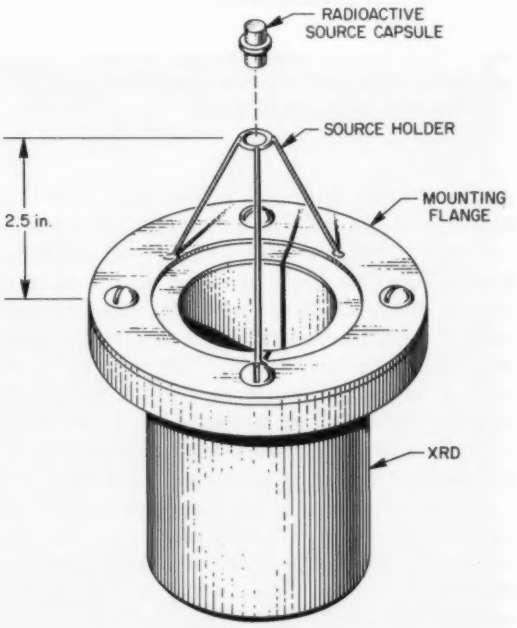

Fig. 2 Suggested geometry for mounting a radioisotope source used in detector calibration.

Energy fluxes from the radioisotopes can be measured by either counting or dosimetry techniques and the results compared and correlated with the predicted and measured source strengths. Any discrepancies outside the experimental error must be resolved. Time-decay corrections to energy flux numbers must be made for short-lived isotopes.

Detector output currents in the 10^{-12}- to 10^{-13}-A range will be measured using vibrating-reed electrometers, as is now done in calibrations with X-ray machine sources. Spectral purity can be demonstrated through spectral measurements with NaI and Ge(Li) detectors and comparison of measured attenuation coefficients with accepted values of this parameter. (MG)

REFERENCES

1. C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes*, 6th ed., John Wiley & Sons, Inc., New York, 1968.
2. R. E. Lewis and T. A. Butler, Reactor Production and Characterization of ^{109}Cd, *Isotop. Radiat. Technol.*, 6(1): 41-43 (Fall 1968) (ORNL-4247).
3. R. E. Lewis, Oak Ridge National Laboratory, personal communication, August 1968.
4. J. H. Gillette, Review of Radioisotopes Program, 1965, USAEC Report ORNL-4013, Oak Ridge National Laboratory, 1966.
5. W. S. Emmerich, S. E. Singer, and J. D. Kurbatov, Radiative Electron Capture of ^{55}Fe, ^{37}A, and ^{59}Ni, *Phys. Rev.*, 94: 113-116 (1954).
6. D. T. Goldman and J. R. Roesser, *Chart of the Nuclides*, 9th ed., General Electric Company, Schenectady, N. Y. 1966.
7. C. Foin, A. Gizon, and J. Oms, Disintegration of ^{109}Cd→^{109}Ag (in French), *Nucl. Phys.*, A113: 241-248 (1968).

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Gamma-Ray Weld-Inspection System*

By D. M. Payne†

Abstract: *A system containing three ^{170}Tm gamma-radiation sources was used to inspect an opposed-arc weld for weld-nugget penetration in an Al–Cu alloy. The system compared the difference between gamma-radiation attenuation through the center of the weld, where attenuation was greater owing to concentration of copper, with that through the sides. The amount of copper concentration increased with weld–nugget overlap.*

A gamma-ray method for inspecting the opposed-arc weld—one of the more difficult welds to inspect—is described here.

The method is based on the principle that, for a constant weld thickness, the attenuation (I) of a gamma-ray beam (I_0) passing through the weld is a function of the density of the material that makes up the weld:

$$I = I_0 e^{\mu_m \rho t}$$

where μ_m, ρ, and t are the mass absorption coefficient, density, and thickness, respectively, of the weld material. If the weld material contains components that segregate on freezing, the resulting concentration can be detected if there is a significant difference in the absorption coefficients of the concentrate and matrix.

The three possible arrangements of the weld material are nugget penetration, nuggets just touching, and nuggets separated by a void space (Fig. 1). Only the first condition gives an acceptable weld. Our work was done with $^5/_8$-in.-thick aluminum (2014 alloy) plates welded with aluminum (2219) rod. The most important alloying element—copper—is significantly denser than aluminum, and it tended to concentrate in the last metal to solidify. Thus, in condition 1, the copper density would be greatest in the double-cross-hatched region. It would be less in condition 2 than in condition 1, and even less in condition 3 because of the void space.

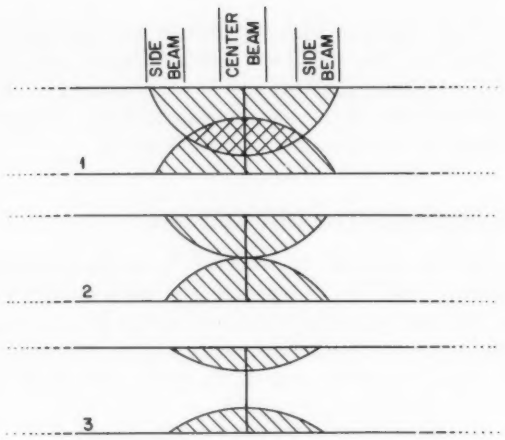

Fig. 1 Three types of welds: (1) nugget penetration, (2) nuggets just touching, and (3) nuggets separated by a void space. Only condition 1 is an acceptable weld.

WELD-INSPECTION SYSTEM

A weld-inspection system (Fig. 2) was developed which makes use of gamma attenuation by weld material to determine whether or not a weld is acceptable. The inspection head of a scanner contains three ^{170}Tm sources (12 mCi each) (^{170}Tm maximum gamma energy = 0.084 MeV). The head (Fig. 3) is passed along the weld to be inspected at a rate of 1 in./min so that one center and two side collimated beams ($^1/_8$ by 1 in. cross section) pass through the weld. On the other side of the weld the beams are again collimated, and the center beam is detected by one gas proportional counter and the two side beams by another. The top collimator is essential because the radiation mechanism in the aluminum is exclusively Compton scattering, in which the gamma photons are deflected and lose energy but are not absorbed.

The center beam indicates the change in density of the weld material due to weld penetration. The two side beams indicate weld-thickness variations. The signals from the two detectors are amplified by two preamplifiers mounted on the inspection head.

In the console the center and side signals are again amplified. Since the signal from the two side beams gives a count rate approximately twice that from the center beam, the side-beam signal is divided by 2, and

*The system was constructed for the National Aeronautics and Space Administration.

†Atomics International Division, North American Rockwell Corp., Canoga Park, Calif.

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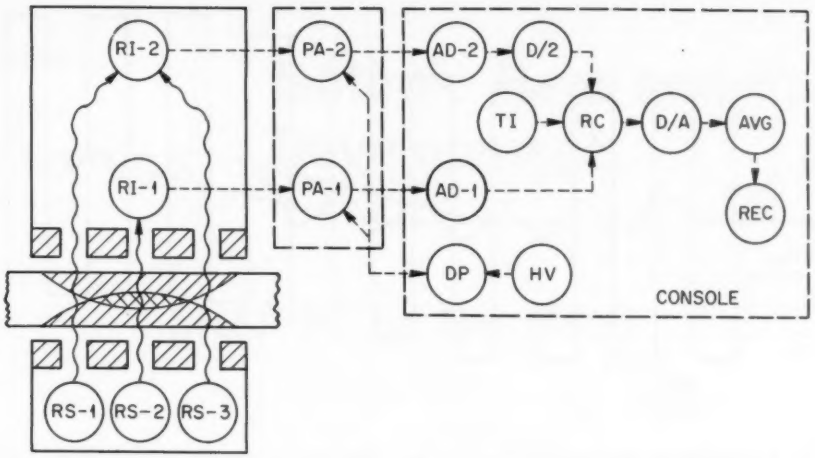

Fig. 2 Weld-inspection system. RS-1, -2, -3 = ^{170}Tm Sources; RI-1, -2 = proportional counters; PA-1, -2 = preamplifiers; AD-1, -2 = amplifier discriminators; D/2 = divider; TI = timer; RC = reversible counter; D/A = digital-to-analog converter; AVG = averager; DP = distribution panel; REC = recorder; HV = high-voltage supply.

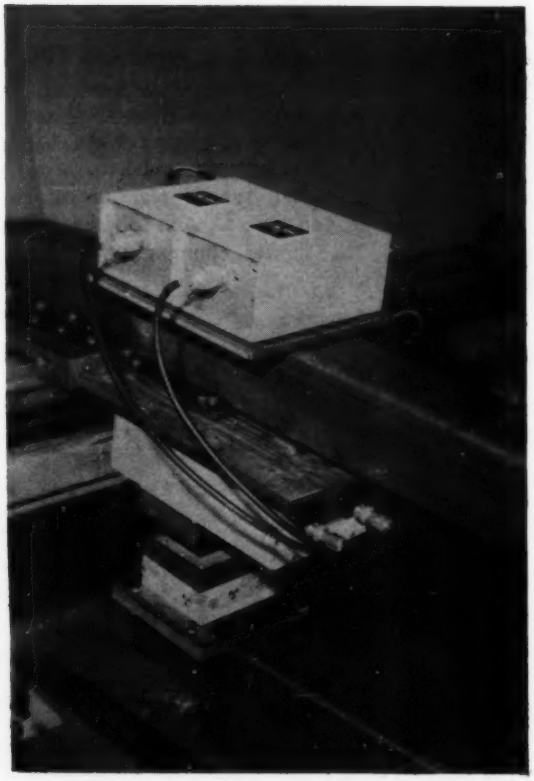

Fig. 3 Inspection head.

then the two signals are simultaneously subtracted from each other in the reversing counter for an interval determined by the timer. The digital output of the counter is then converted to an analog signal.

Owing to statistical variations in the signals, the analog signal varies considerably from one counting interval to the next, even during the traverse of metal of constant density. Therefore an averaging circuit is included to smooth out the variation, and the smoothed signal is recorded. The averaging circuit consists of an operation amplifier with a capacitor paralleled by a resistor in the feedback loop. The time constant is 3 min for a 20-sec counting interval. In Fig. 4, with an output signal of a constant 1-V amplitude and a 1-min pulse width, the output averaging-circuit trace on the right would be that from a condition 1 weld, i.e., an acceptable weld. The sloping part is a transition through a condition 2 to a condition 3 weld. An actual recorder trace shown in Fig. 5 indicates a condition 1 weld at the bottom. After about three squares the weld goes through conditions 2 and 3.

As shown in Fig. 4, the voltage rises as the capacitor charges and falls as it discharges, and the circuit works because the voltage varies much faster as a result of the statistical variation than it does as a result of weld changes. The disadvantage of using this circuit is shown in the central part of Fig. 4 where the averaging-circuit output lags behind the weld changes.

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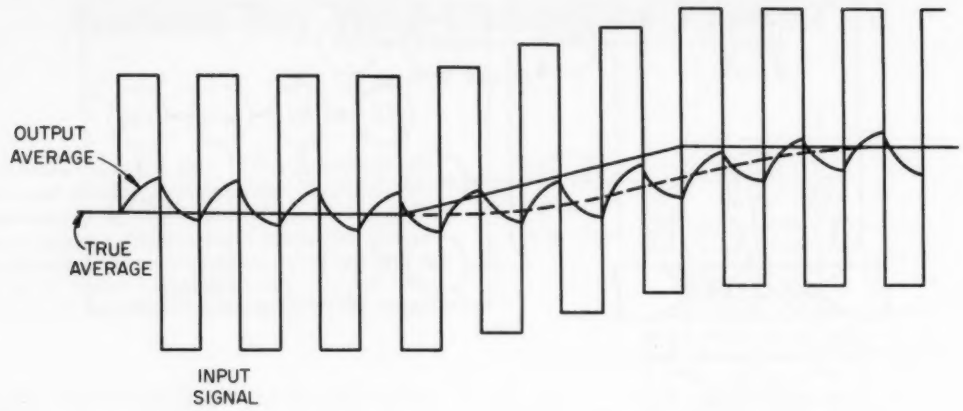

Fig. 4 Averaging-circuit output for a constant 1-V amplitude and a 1-min pulse-width input signal.

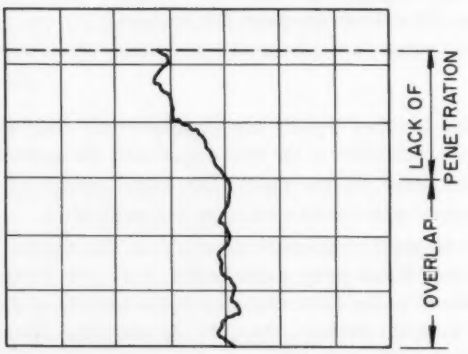

Fig. 5 Recorder trace.

In this system, where the inspection head is approximately 3 in. beyond the weld parameter change, the problem could be compensated by measuring 3 in. back from the apparent to the actual parameter change point. The use of a shorter counting time, which would result in a decrease in the weld signal, necessitates decreasing the inspection-head scanning speed. However, even a 1-min counting time is not acceptable in practice, and a more feasible alternative would be to replace the ^{170}Tm sources with some radioisotope that gives a larger number of gamma photons in the 0.08-MeV energy region, e.g., ^{241}Am, to shorten the counting time. The shorter counting time would produce a smoother curve. (MG)

Section III

Isotopes and Radiation Technology

Isotopic Heat and Power Development

SNAP-27 on the Moon

By D. L. Prosser*

Abstract: *A SNAP-27 thermoelectric generator left on the moon by the Apollo 12 astronauts was designed to provide electrical power for five experiments on the lunar surface. The source is fueled with 3735 g (8.36 lb) of ^{238}Pu dioxide microspheres, which produce 63 W(e) [1480 W(th)] of power. The sources were designed, fabricated, encapsulated, and extensively tested to ensure the integrity of the system prior to flight authorization.*

The SNAP-27 system† was designed to provide power for the Apollo Lunar Surface Experiments Package (ALSEP) developed for the NASA Manned Spacecraft Center. The ALSEP, a package of instruments and supporting subsystems, was placed on the lunar surface (Fig. 1) by a crew member of the lunar module (LM) vehicle and is expected to transmit lunar environmental information for at least a year. The equipment in the package is

1. Passive Seismic Experiment: to measure lunar seismic activity and detect meteoroid impact and free oscillations in lunar rotation and possibly surface tilt tidal deformations resulting, in part, from periodic variations in the strength and direction of external gravitational fields acting on the moon.

2. Magnetometer Experiment: to measure the magnetic flux of the moon and possible effect on the moon of other magnetic fields in the solar system.

3. Solar Wind-Detector Experiment: to measure the ion flux on the moon resulting from solar activity.

4. Supra-Thermal Ion-Detector Experiment: to measure protons and ions other than those measured by the Solar Wind Detector.

5. Cold Cathode-Gage Experiment: to measure small gas pressures and the low density of the lunar atmosphere.

Six sources were fabricated for the SNAP-27 program—one for test purposes, four for flight missions, and one as a stand-in for these four. The first of these four sources was transported to the moon by the Apollo 12 vehicle.

POWER SOURCE

Each power source (Fig. 2, Table 1) is fueled with about 3735 g (8.36 lb) of ^{238}Pu dioxide microspheres to produce 63 W(e) [1480 ±30 W(th)]. The half-life of ^{238}Pu is 87.4 years; it decays predominantly by alpha-particle emission, with small gamma and neutron radiation. The resultant power decay will be less than 1% per year.

The thermal-power requirement was achieved with 50- to 250-μm-diameter $^{238}PuO_2$ microspheres prepared by fusion of oxide particles in a plasma torch. Properties of the fuel were

Apparent density	10.08 to 10.54 g/cm^3
Total impurities	0.26 to 0.94 wt. %
Stoichiometry	2.00
^{238}Pu content	70.3 to 71.3 wt. %
Melting point	2255 ± 90°C
Fuel per capsule	~3735 g (~44,500 Ci) (8.36 lb)

Electricity is generated as the decaying radioisotope heats a series of tiny thermocouples (Fig. 3), which are connected in series around the radius of the fuel chamber.

*Mound Laboratory, operated by Monsanto Research Corp. for the USAEC. More detail is given in "SNAP-27 Radioisotopic Heat Source Summary Report, USAEC Report MLM-1698, Mound Laboratory, Nov. 11, 1969."

†The power source was designed and the components provided by General Electric, and the fuel was fabricated and encapsulated by Mound Laboratory. The 3M Company fabricated the thermocouples and assembled the generator components except for the fuel element and cask.

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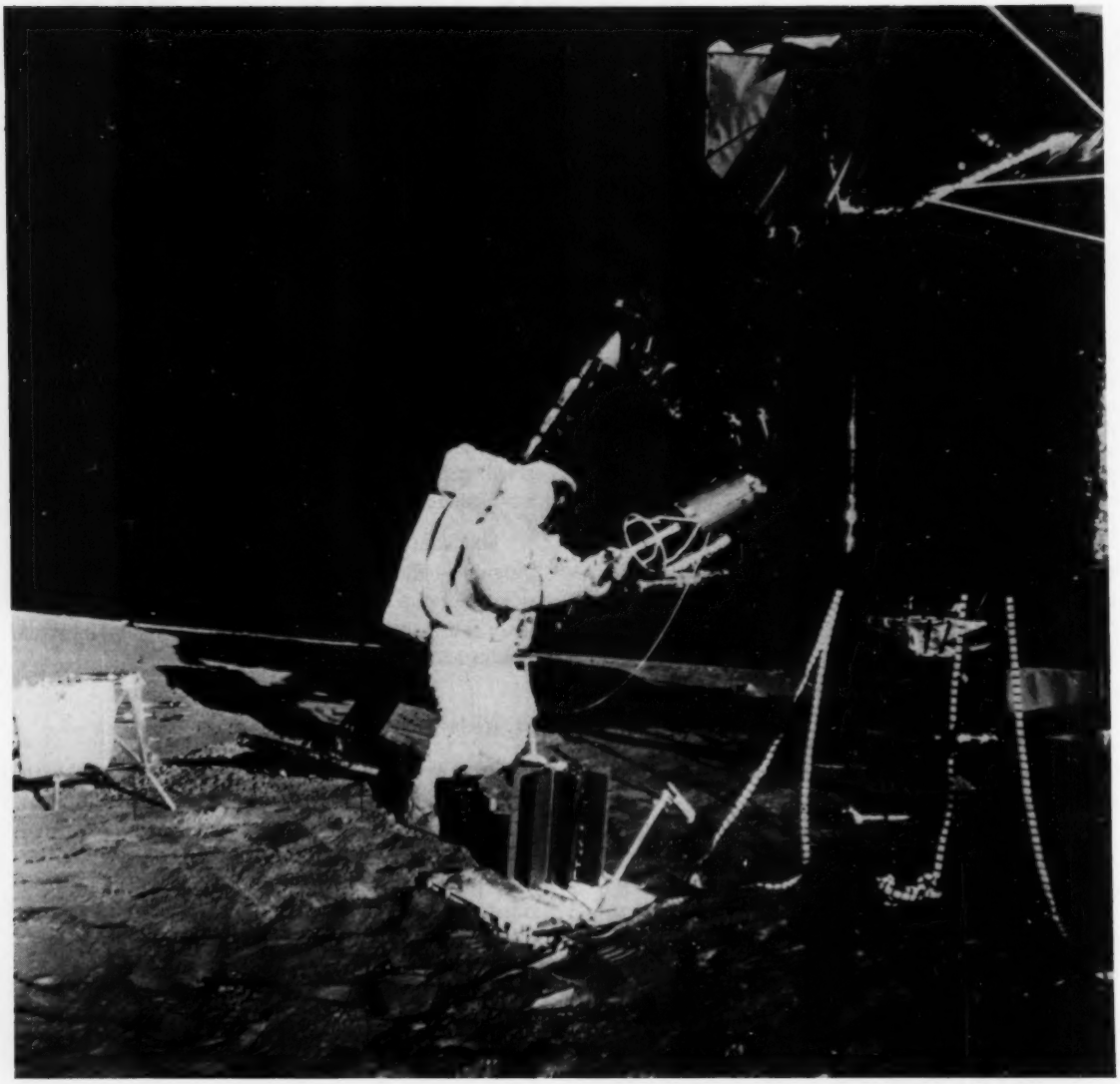

Fig. 1 Astronaut Allen L. Bean pulls the radioisotope fuel element from its carrying cask for insertion in the SNAP-27 thermoelectric generator on the moon. (Photograph courtesy of NASA.)

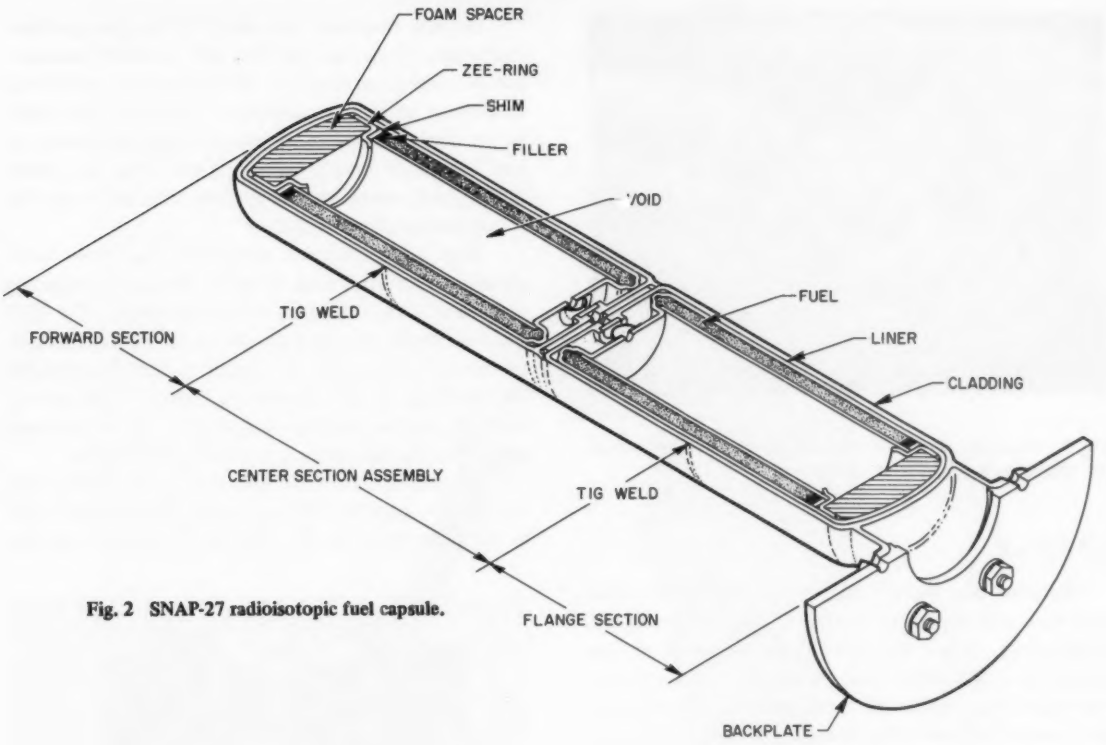

Fig. 2 SNAP-27 radioisotopic fuel capsule.

Table 1 Design Parameters for SNAP-27 Heat Source

Thermal loading, W	1480 ± 30
Fuel	
Form	$^{238}PuO_2$ microspheres, 50 to 250-μm diameter
Geometry	Cylindrical annulus
Specific power, W/g	0.400 ± 0.01
Particle physical density, g/cm^3	9.1 to 10.3
Effective power density, W/cm^3	2.6 + 0.2 − 0.1
Effective thermal conductivity in 98% helium at 1400°F cladding temperature, Btu/(hr)/(ft)/(°F)	0.62
Capsule	
Fueled length, in.	13.76
Fueled length, cm	34.95
Outside diameter (nominal, uncoated), in.	2.509
Outside diameter (nominal, uncoated), cm	6.373
Cladding material	Haynes alloy No. 25
Emissive coating	Radifrax RC-356
Emissivity	0.85 (minimum)
Mission time, years	1
Storage time, years	2
Total weight (with backplate), lb	15.46 maximum
Total weight (with backplate), kg	7.03 maximum

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Fig. 3 Thermocouples used in generating electricity in SNAP-27. (Photograph courtesy of 3M Company.)

CAPSULE

Because this source contained considerably more fuel than any source fabricated to date for aerospace applications, major importance was attached to the design of a heat-protection system that would permit the capsule to survive an abort from earth orbit and land intact without dispersing plutonium within the biosphere.

The capsule is of a dual design, each half having an annular liner that holds the fuel. Helium gas generated in decay of the fuel accumulates in the inner volume of the liner to minimize pressure buildup in the capsule. Each half capsule has a venting device to release the helium gas before the pressure reaches a point that might jeopardize the integrity of the source. All seals were made by either electron-beam or tungsten-inert-gas (TIG) welding methods. The outer cladding (Table 1) was fabricated from Haynes alloy No. 25, used because of its high strength and resistance to oxidation at the operating temperature of the source.

The fuel was sealed in the annular cavity of each of the two liners. Both liners were then rigorously tested to ensure integrity of the seal, and the thermal output of each liner was determined. One liner was placed in the prewelded center-section assembly (Fig. 2), the forward section was placed over the exposed end of the liner, and the two cladding components were joined by TIG welding. The second liner was enclosed in the other end of the center-section assembly by using the flange-section component. The welds were examined closely to be sure of their integrity. The cladding assembly provides a capsule free of surface radioactive contamination and acts as a strength member.

The test capsules were subjected to a comprehensive series of mechanical tests and radiation measurements, being exposed to all conceivable accidental conditions of impact, explosion, and heat that might be encountered before, during, or after the launch of the Saturn V space vehicle. Only after all safety requirements were proved to have been met were the sources authorized for flight.

After all inspection operations had been completed, the sources were stored in the ground shipping cask (GSC) until shipped to Cape Kennedy. The GSC was specifically designed to attenuate the neutron and gamma radiation from the capsule to a level acceptable for handling. It also provided a system to adequately cool the capsule during storage or transportation and against accidental handling and shipping hazards.

For transportation to the moon, the source was placed in a graphite LM fuel cask (GLFC) rather than in the generator (Fig. 4). The GLFC provided protection for the source in the event of a possible launch-pad explosion and also protected the source against heat of reentry in the case of an abort from space.

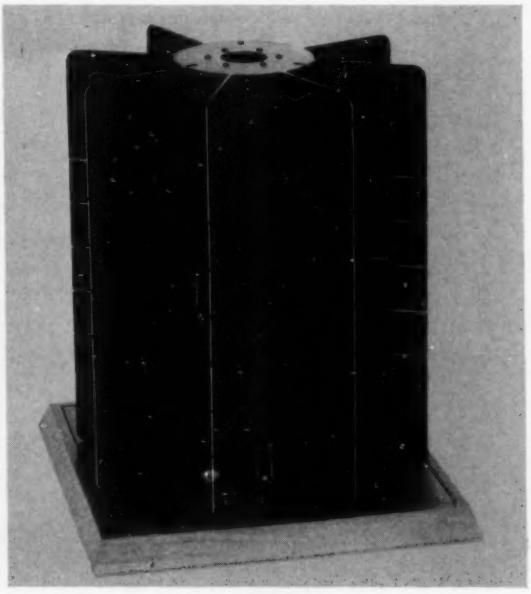

Fig. 4 SNAP-27 generator assembly. (Photograph courtesy of 3M Company.)

Upon reaching the moon, the source was removed from the GLFC with a special handling tool (Fig. 5)

Fig. 5 Demonstration of loading, using special handle, of fuel capsule into full-scale model of SNAP-27 generator. (Photograph courtesy of 3M Company.)

and placed in the generator. This tool was designed to be locked to a backplate attached to the flanged end of the heat source. The backplate was also used for locking the source into either the GLFC or generator.

* * *

Preliminary information indicated that the radioisotopic generator was continuing to operate 1 month after it was placed on the moon's surface on November 19. During the first 28-day lunar day-and-night cycle, temperatures ranged from −291 to +283°F. Powered by the more than 70 W of electricity being produced by the generator, the five instruments left on the moon's surface by the astronauts of Apollo 12 were sending back lunar scientific data.

(MG)

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Radioisotopic Heat Source on the Moon

By D. P. Kelly and V. L. Avona*

Abstract: *Two radioisotope-fueled heaters were left on the moon by the Apollo 11 astronauts for protection of seismic instrumentation. Each heater was fueled with 37.6 g of $^{238}PuO_2$ microspheres, which produce 15 W(t). The sources were designed, fabricated, encapsulated, and extensively tested to ensure integrity of the system prior to flight authorization.*

When Apollo 11 astronauts landed on the moon on July 21, 1969, they deposited on the moon's surface the Early Apollo Scientific Experiments Package (EASEP). This package contained two Apollo Lunar Radioisotopic Heaters (ALRH) (Fig. 1) to keep the electronic seismic instrumentation at temperatures of −65°F (−54°C) or higher during the lunar night, when temperatures may reach −300°F (−184°C). During the lunar day, insulation and a reflective coating protect the package from excessive heat, maintaining the temperature at 0 to 140°F (−18 to 60°C). The package also contains the Laser Ranging Retro-Reflector (LRRR).

Because only 6 months was available for the work, a philosophy of overdesign and overtesting was adopted. Three fueled heaters were fabricated, one as backup if needed.

POWER SOURCE

The thermal-power requirement of 15 ± 0.25 W(t) for each ALRH was met by using 37.6 g of $^{238}PuO_2$ microspheres, 50 to 250 μm in diameter. The fuel properties were

Apparent density	10.14 g/cm^3
Total impurities	<0.44 wt.%
Stoichiometry	2.00
^{238}Pu content	70.7 wt.%
Melting point	2370 ± 80°C
Source strength	~450 Ci

*Mound Laboratory, operated by Monsanto Research Corp. for the USAEC. More details are given in "Apollo Lunar Radioisotopic Heater Summary Report," USAEC Report MLM-1637, Mound Laboratory, Apr. 30, 1969.

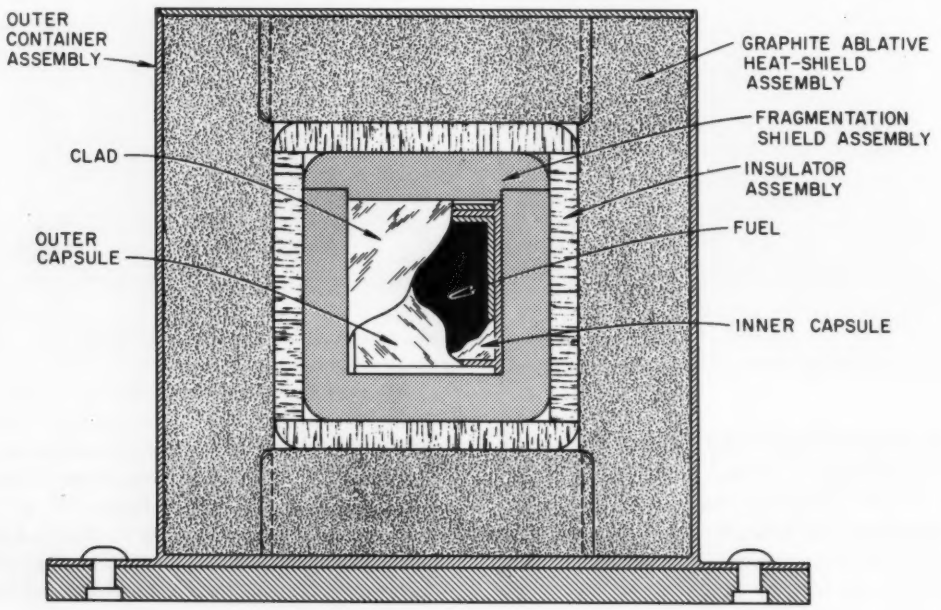

Fig. 1 Cross section of the ALRH.

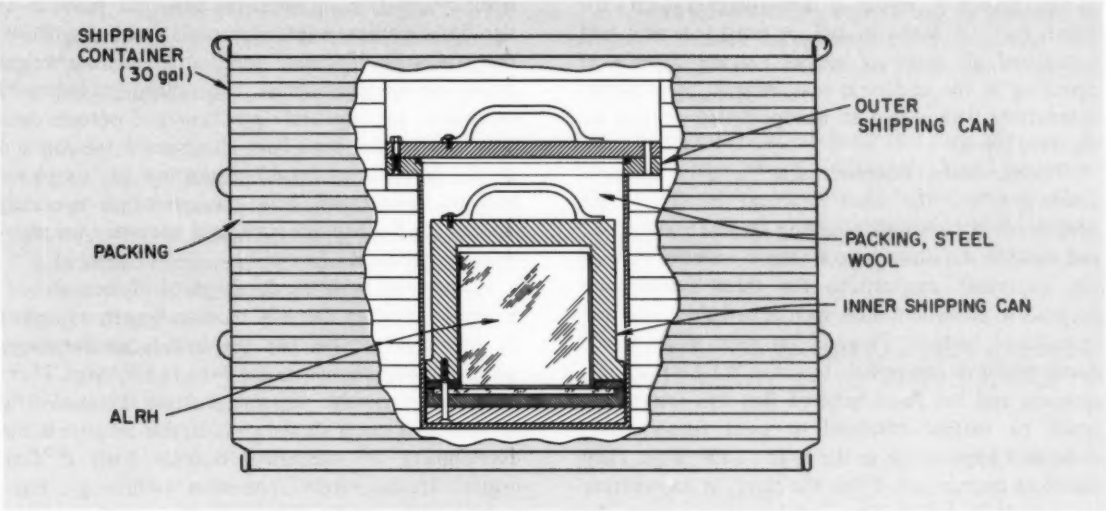

Fig. 2 Shipping container for the ALRH.

CAPSULES

The fuel was doubly encapsulated. The inner capsule—0.785 in. (1.994 cm) in inside diameter, 0.825 in. (2.096 cm) in outside diameter, and 8.22 in. (2.088 cm) long—was made of Ta–10 wt.% W refractory alloy, which is compatible with the fuel. A 20-mil (0.051 cm) thickness of this material is not difficult to weld by the tungsten–inert gas (TIG) technique.

The outer capsule, of the same material and wall thickness, was 0.835 in. (2.121 cm) in inside diameter, 0.875 in. (2.22 cm) in outside diameter, and 0.886 in. (2.250 cm) long. As an oxidation barrier for the refractory metal, a 10-mil-thick cladding of Pt–10 wt.% Rh was TIG-welded to the outer surface of this second capsule.

The fuel-capsule assembly was surrounded by a fragmentation shield for mechanical protection against a possible launch-pad explosion or terminal-velocity impact; a high-temperature insulator as protection in case of an abort from space; and a graphite heat shield. All were enclosed in a 304 stainless-steel container, which was riveted to a base plate.

The assemblies, with simulated fuel elements, were extensively tested, and each was shipped to the launch site in an individual container (Fig. 2). (MG)

Isotopic Energy Systems for Underseas Application*

Summarized by Martha Gerrard

Interest in underwater operations has generated a variety of mission requirements with a wide range of energy needs. Because of the Navy's urgent need for a comprehensive program for developing energy systems for underseas use, the Conference on Energy Sources of Extended Endurance in the 1–100 kW Range for Naval Applications was held at the National Academy of Sciences, Washington, D. C., in April 1967. The Committee on Undersea Warfare and the Office of Naval Research sponsored the conference, and the Committee subsequently assembled a group of experts as its Panel on Energy Sources. Under the chairmanship

*Extracts from A Report by the Panel on Energy Sources of the Committee on Undersea Warfare, National Research Council, Publication 1702, National Academy of Sciences, Washington, D. C., 1968 (edited by R. M. Chapman, D. A. Patterson, et al.).

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of Dr. Howard A. Wilcox of Santa Barbara, Calif., the Panel met 14 times in full or partial session and considered all types of energy systems capable of operating in the underseas environment. Their recommendations with regard to isotope-fueled systems are reported below.

Isotope-fueled systems, using static or dynamic converters, are the most practical candidates for deep-ocean applications requiring up to 15 kW power and mission durations longer than 6 months. Among the important applications for these systems are deep-ocean acoustic devices such as navigation aids and surveillance systems. Progress has been made in the development of low-power (less than 0.1 kW) isotopic systems, and the Panel believed that this technology could be further advanced to meet future, long-endurance applications in the 1- to 15-kW range. They therefore recommended that the Navy, in conjunction with the U. S. Atomic Energy Commission, pursue the development of a family of isotopic power systems on a priority basis, stressing improved conversion efficiency, reliability, and cost effectiveness. The systems should be developed on a modularized design basis and should be qualified for use in a maximum depth environment.

Isotope-fueled systems have a number of advantages for use in the ocean environment. They are compatible with an underseas environment, and they do not require frequent maintenance. Up to 15 kW or more of electric power may be obtained from them, and they are available as compact, self-contained units. Their safe operation in the ocean environment has been demonstrated.

The Panel felt that the increasing recognition of isotopic power as a potentially attractive energy source for remote environments requiring continuous and unattended operation for a year or more is largely the result of the SNAP-7 program, completed in 1966, which demonstrated the capability of isotopic devices for long-term, unattended, safe, and reliable operation in a variety of hostile environments. Isotopic power at present [at the time of the Panel meeting] has been limited to less than 0.1 kW in thermoelectric devices using ^{90}Sr as a heat source. The devices have been relatively heavy and inefficient—power/weight ratio of 10^{-2} W/lb and end-of-life efficiency of 3 to 4%. Operating temperatures have been generally restricted to 800 to 900°C, primarily because of limitations of engineering materials.

The AEC has now significantly expanded its research and development activities, and second-generation devices (SNAP-21 and -23) are under development (1967). These sources offer promise for significant improvements in isotopic fuel management, thermoelectric efficiency and stability, power/weight ratios, operating lifetimes, and reliability. Improved techniques for large-scale production of isotopic fuels at relatively low costs have also been developed and should lead to a second generation of low-power isotopic devices that have the reliability, operating lifetime, and economic advantages necessary for widespread application.

Concurrent with the development of these second-generation devices, the AEC has significantly expanded its programs and has laid the foundation for larger isotopic power sources in the 1- to 15-kW range. There have been noticeable improvements in thermoelectric properties of materials and considerable progress in the development of alternative isotopic fuels (^{60}Co), organic Rankine-cycle conversion technology, high-temperature insulation materials, fuel production and encapsulation techniques, and radiological safety design criterions. Additional programs being pursued in other government agencies (NASA, Brayton cycle) and industrial laboratories (General Motors, Stirling engine; Battelle–Northwest, radioisotope Stirling) should materially enhance the capability of developing large isotopic systems in the power range of interest to the Panel.

In recognition of the potential need for large isotopic power sources in the range up to 15 kW, the AEC has implemented engineering design and applications studies at Oak Ridge National Laboratory. Initial design criterions relating to these studies are

Power Level:	1 to 15 kW
Operating lifetime:	3 months to 5 years
Isotopic fuel:	^{60}Co, ^{90}Sr, and ^{144}Ce
Power conversion:	Thermoelectric, Brayton engine, organic Rankine engine, liquid-metal Rankine engine, and Stirling engine
Environment:	Seawater; 20,000-ft depth at 50°F

The primary objectives of these studies are to (1) evaluate parametrically alternative energy-conversion cycles within the restrictions imposed by realistic isotopic fuel-block designs; (2) determine isotopic fuel and shielding requirements and heat-exchanger design requirements; (3) design alternative emergency cooling provisions; and (4) establish performance capability and cost-effectiveness criterions. The upper power limit of 10 to 15 kW to be studied does not represent a maximum projected power capability, but rather is predicated on the availability of isotopic fuels over the next 5 to 7 years. Further

applications requiring power sources in the 20- to 50-kW range may be satisfied with either isotopic or reactor systems, although neither system appears to offer clear-cut advantages over the other in this power range.

Although these studies have begun only recently, the Panel, through its discussions with the AEC, was able to identify several areas that will require special attention during the follow-on development effort:

1. Increased thermoelectric conversion efficiencies.
2. Improved reliability in the design and operation of small rotating machinery involving degradation of working fluids, gas bearings, and seals.
3. Development of high-temperature (>1000°C) ^{60}Co encapsulation techniques.
4. Determination of engineering safeguards to prevent fuel-block damage resulting from coolant loss.
5. Demonstration of low-cost production capability for selected isotopic fuels.

It is important to recognize that these advances, as well as others previously discussed, should be attainable by an orderly research and development effort without the necessity for unpredictable technological breakthroughs.

* * *

The report also discusses some other systems and ancillary equipment and has a bibliography of 50 items.

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Section
IV
Isotopes and Radiation Technology

Isotopes Around the World

Radioisotope Program in India

By V. K. Iya* and R. G. Deshpande*

The radioisotope program in India was started soon after the commissioning of the first swimming-pool reactor, APSARA.[1] The initial purpose of this program was service to isotope users, mainly researchers, who were then importing isotopes. With the commissioning of the 40-MW CIRUS in 1960 and installation of handling facilities at the Intermediate Isotope Laboratory at Trombay in 1961, the scope of the production program broadened. More radioactive products were gradually produced, and their application in medicine and industry was actively encouraged.[2,3] By late 1969, the initial aims of the isotope program were considered to have been fulfilled. About 350 radioactive products are available from the Isotope Division, and these have proved to be valuable tools for Indian work in medicine, agriculture, industry, and hydrology. During 1968, about 18 thousand consignments of radioisotopes were supplied to 325 different institutions, with a total sales value of Rs. 2.2 million (U. S. $300 thousand).

Further expansion of the production and application programs is envisaged with the multikilocurie handling and development capabilities in the new radiological laboratories at Trombay (Fig. 1). These laboratories provide extensive radiochemical facilities for processing a wide variety of radioactive materials at multicurie levels. For isotope production, an area of approximately 34 thousand ft^2, including laboratory, office, and service arrangements, is available, in addition to two hot cells designed to handle 10 kCi of ^{60}Co. The power reactors under construction at Rajasthan and Madras would allow production of large quantities of various isotopes—in particular, megacurie quantities of ^{60}Co. The High Intensity Radiation Utilisation Project, now being undertaken, will enable the development of high-intensity-radiation applications in the near future.

There has been a steady growth in the application of radioisotopes in India (Table 1). Currently, medical and industrial uses each account for 35% of the total radioisotope sales, with research and agriculture together accounting for the remaining 30%. The value of exports is approximately 12% of the total sales.

RADIOISOTOPE PRODUCTION

The radioisotope products made at Trombay are primary isotopes, labeled compounds—radiopharmaceuticals, biochemicals, organic and inorganic compounds—and sealed radiation sources. Radioisotopes are produced by (n,γ), (n,p), and (n,f) reactions, the targets being irradiated in either APSARA (1-MW swimming-pool reactor) or CIRUS (40-MW, heavy-water-moderated, natural-uranium-fueled tank reactor). APSARA offers a maximum flux of about 3×10^{12} $n/(cm^2)(sec)$, 30% of which is fast flux. The maximum thermal neutron flux in CIRUS is 6×10^{13} $n/(cm^2)(sec)$.

Extremely pure targets are used. In some cases, to increase the specific activity of the desired radioisotope, the targets are reprocessed. For example, irradiated potassium chloride is recovered after the separation of carrier-free ^{35}S, recrystallized, dried, and then reirradiated to increase the specific activity of the ^{36}Cl produced in the target by the $^{35}Cl(n,\gamma)^{36}Cl$ reaction. Electromagnetically enriched targets are also routinely used for producing ^{54}Mn and ^{59}Fe, for example. In the former case, after separation of ^{54}Mn, the enriched iron target is recovered and reirradiated. Recoil enrich-

*Isotope Division, Bhabha Atomic Research Centre, Trombay, Bombay-85(AS), India.

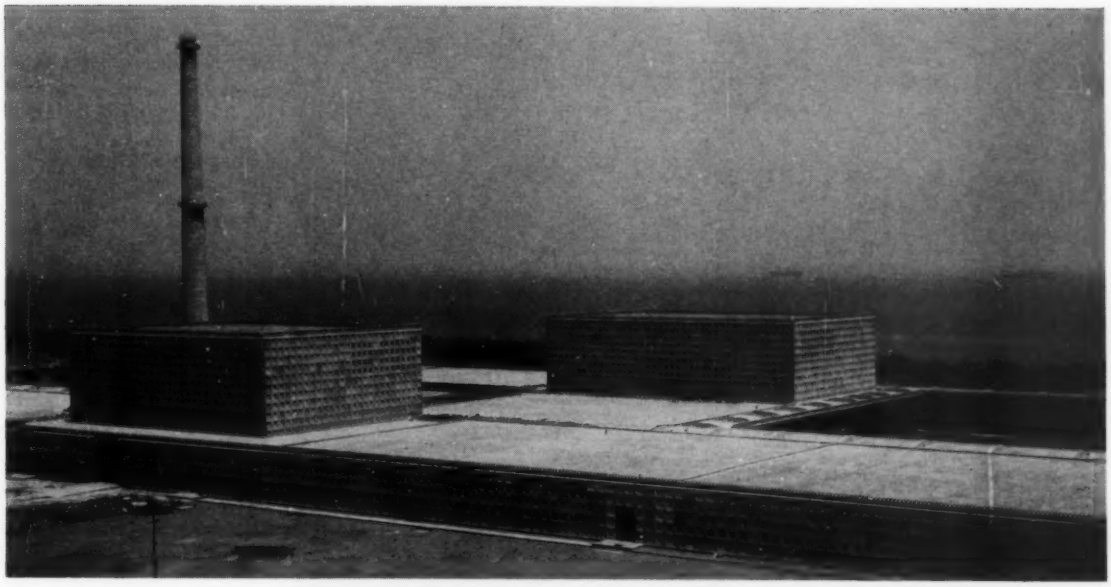

Fig. 1 New radiological laboratories at Trombay.

Table 1 Growth of Radioisotope Uses in India

Year	No. of users	No. of consign-ments	Sale value,* Rs.
1958	8	40	600
1959	15	75	1,200
1960	35	360	15,000
1961	58	761	48,600
1962	82	1,490	361,550
1963	126	2,914	309,993
1964	151	3,473	632,948
1965	196	3,959	832,326
1966	264	7,997	1,023,658
1967	309	10,945	1,372,069
1968	325	18,000	2,200,000

*1 rupee = U. S. $0.133.

ment is used for production of some high-specific-activity isotopes, e.g., ^{51}Cr.

Millicurie quantities of fission products of short and medium half-lives—e.g., ^{97}Zr, ^{95}Zr, ^{95}Nb, ^{143}Ce, ^{140}Ba, ^{99}Mo, and ^{132}Te— are separated from irradiated uranium targets.

A program for recovery of kilocurie quantities of long-lived fission products—^{137}Cs, ^{90}Sr, and ^{147}Pm—from the reactor-fuel processing plant effluent at Trombay has been proposed. Laboratory studies on separation using inorganic ion exchangers are being conducted. Curie amounts of these isotopes will be produced in the new radiological laboratories.

The production methods followed at Trombay for processing radioisotopes are described in detail in "The Manual of Radioisotope Production," IAEA, 1966. Each method was selected to take advantage of locally available materials and equipment.

Primary Radioisotopes

More than 50 different radioisotopes are regularly produced (Fig. 2), the most important of which are

Isotope	Present annual production capacity, Ci	Isotope	Present annual production capacity, Ci
^{131}I	200	^{51}Cr	6
^{32}P	60	^{58}Co	1.5
^{198}Au	100	^{203}Hg	15
^{35}S	10		

Others include ^{45}Ca, ^{63}Ni, ^{99}Mo, ^{132}Te, and ^{140}Ba. Molybdenum-99 and ^{132}Te are also supplied in the form of generators from which the very-short-lived ^{99m}Tc and ^{132}I, respectively, can be eluted.

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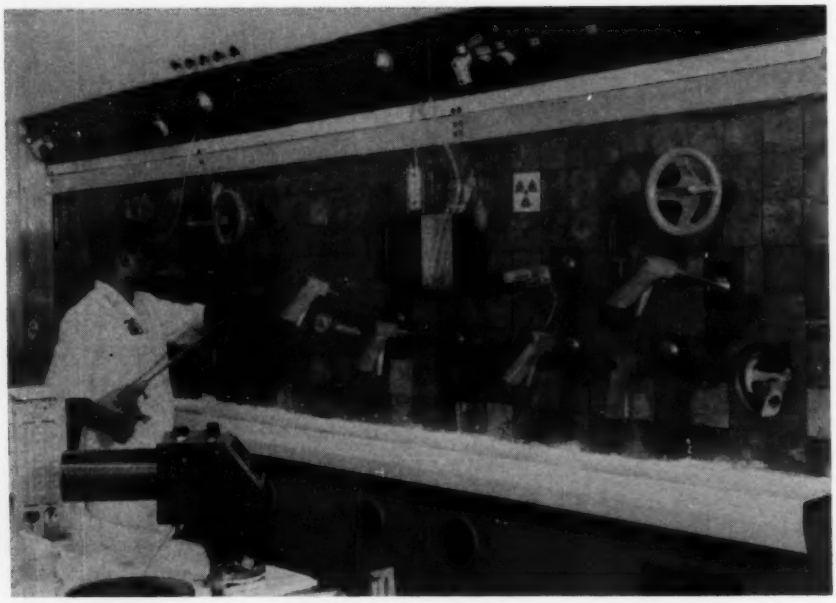

Fig. 2 Shielded enclosure for processing radioisotopes.

Labeled Compounds

Radiopharmaceuticals. Specifications have been standardized for about 55 radiopharmaceuticals, which are produced regularly.[4] The products include several ^{131}I- and ^{125}I-labeled compounds, e.g., Rose Bengal, triolein, oleic acid, Hippuran, and RISHA; ^{197}Hg- and ^{203}Hg-labeled Neohydrin; and ^{57}Co- and ^{58}Co-labeled vitamin B_{12}. Other radioisotopes involved include ^{32}P, ^{198}Au, ^{51}Cr, ^{24}Na, ^{42}K, and ^{82}Br.

All starting materials for production of radiopharmaceuticals are of the highest purity, and sterile, pyrogen-free water is used. These compounds are prepared under aseptic conditions (Fig. 3) in production plants fitted with ultraviolet germicidal lamps, and a sterile atmosphere is maintained in the production laboratory. All radiopharmaceutical products are subjected to physical, chemical, radiochemical, and biological tests[5] prior to sale to ensure that they conform to specifications. Although radiopharmaceutical production in India is a monopoly of the Isotope Division of Bhabha Atomic Research Centre, these products are released for medical use in accord with the Directorate of Drugs Control Administration standards. To ensure maintenance of the highest safety standards, the Radiopharmaceuticals Committee examines and approves all procedures. A Nuclear Medicine Committee has also been constituted to license individual medical users. As new radiopharmaceuticals are developed, they will be introduced into the regular supply schedule only after approval of both the Radiopharmaceuticals Committee and the Nuclear Medicine Committee.

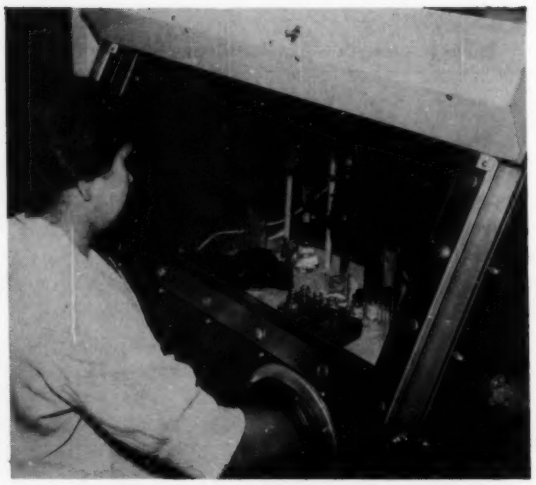

Fig. 3 Aseptic glove box for preparation of radiopharmaceuticals.

Biochemicals. A hundred or more ^{14}C- or ^{3}H-labeled compounds are prepared for research purposes, including amino acids, sugars, nucleotides, carboxylic acids, pyridines, and purines.

A tritium-labeling service is also offered, which labels compounds supplied by the customer, using appropriate techniques. With the availability of more laboratory space and facilities, the scope of this work will be increased.

Inorganic Compounds. Inorganic compounds labeled with ^{32}P, ^{35}S, ^{82}Br, and ^{46}Sc are supplied. These include fertilizers, e.g., superphosphate-^{32}P; insecticides such as Malathion-^{32}P, Malathion-^{35}S, and Diazion-^{32}P; and industrial tracers, e.g., ^{46}Sc-labeled silt and fly ash of appropriate particle size.

Radiation Sources

A number of sealed radiation sources are made (Fig. 4)—^{60}Co sources for radiography and kilocurie units for gamma irradiators and teletherapy units; ^{192}Ir for radiography; and ^{124}Sb–Be and ^{239}Pu–Be neutron sources.

Cobalt-60. For high-specific-activity requirements, nickel-plated cobalt pellets (1 by 1 mm) are used as targets. Thirty standard irradiation capsules with cobalt pellets distributed in an annular space are irradiated in CIRUS at a position where the maximum flux is 6×10^{13} n/(cm^2)(sec). Low-specific-activity ^{60}Co is produced by irradiation in the graphite reflector of CIRUS at a position where a maximum flux of 10^{13} n/(cm^2)(sec) is available. The target is in the form of 6-by 25-mm slugs jacketed in 1S aluminum, arranged in tiers around an aluminum rod and encased in an aluminum tube. The total annual production of ^{60}Co is 100 thousand Ci.

The High Intensity Radiation Utilisation Project to be commissioned shortly provides for development of large irradiation units for use in research and industry. Construction of a megacurie ^{60}Co handling cell and associated laboratories for physics and chemistry work is envisaged. Study of source and design variables is planned, with actual loading of high-activity ^{60}Co sources in the facilities provided.

Cobalt-60 sources in the form of pencils for use in small irradiators, e.g., the Gamma Chamber–900, are produced regularly. The Gamma Chamber has an irradiation volume of about 1 liter and a maximum dose rate of 5×10^5 R/hr. Similar units with irradiation volumes of up to 5 liters and 20 thousand Ci of ^{60}Co are being designed.

Fig. 4 Fabrication of high-intensity radiation sources.

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Teletherapy sources with a maximum output of 50 rmm in 2-cm-diameter capsules are also supplied. These sources are doubly encapsulated in stainless steel, argon arc-welded, and leak-tested by standard methods.

Iridium-192. Iridium-192 radiography sources are prepared by encapsulating irradiated iridium metal pellets in stainless-steel capsules. Pellets of three different sizes (1.6 by 1.6 mm, 2 by 1 mm, and 3 by 1 mm) are irradiated routinely. Sources of up to 50 Ci activity are fabricated, depending on requirements.

Neutron Sources. Antimony–beryllium neutron sources with an output of up to 1.5×10^7 n/sec are supplied for research purposes. The source is fabricated by enclosing an antimony metal rod in a beryllium shell and encapsulating the assembly in an outer stainless-steel capsule. This capsule is irradiated to the desired strength.

Plutonium–beryllium sources of up to 5 Ci, with an output of about 9×10^6 n/sec, have been fabricated.[6] They are rented to research institutions in India.

Miscellaneous Sources. Light sources are made which incorporate self-luminous systems, e.g., copper-, manganese-, or silver-doped zinc sulfide phosphors activated with ^{147}Pm or ^{3}H. Thallium-204 and ^{210}Po foils are made for thickness gages and static eliminators, and reference sources of a number of radionuclides are regularly supplied for instrument calibration. Deuterated and tritiated zirconium targets for use in fast-neutron generators are under development.

Auxiliary Equipment

In order to encourage the widespread use of radiation sources in industry and research, a program has been undertaken for development of auxiliary equipment needed for use of radioisotopes. Radiography sources are supplied in a shielded unit ready for field use. Units containing up to 30 Ci of ^{192}Ir and 10 Ci of ^{60}Co are regularly supplied to industries for nondestructive testing. Laboratory and field irradiators, shielding containers, and small handling equipment are also provided.

RADIOISOTOPE APPLICATIONS

The Isotope Division is also responsible for the application of radioisotopes in industry. Problems referred by industries are examined carefully, and, wherever considered desirable, appropriate radioisotope techniques are developed to meet the specific need of the industry. The major work of the Applications Group has been in determining silt movement in harbors, and a group has recently been formed for application of radioisotopes in hydrology. However, other important tracer studies include detection of leaks in buried pipes and determination of retention times in chemical reactors. Assistance in installation and maintenance of radioisotope gages is also provided to industries.

Sediment Tracing

Radioisotopes have been used as tracers in studying silt movement in harbors to enable the port authorities to make decisions on dredging. Since 1959, when the first radiotracer investigation was carried out near Bombay Harbor, 24 such investigations have been made in the ports of Bombay, Bhavnagar, Calcutta, Cochin, Karwar, Mormugao, and Visakhapatnam and in the Sethusamudram Canal in the Palk Straits. Techniques developed have made it possible to follow silt movement on the seabed under different seasonal and weather conditions.[7] The most commonly used tracer material is ^{46}Sc-labeled glass for studies lasting 2 to 3 months. An interesting application has been in the study of silt movement in the Sethusamudram Canal project, where the results confirmed that the proposed canal alignment was advisable.[8] The proposed canal will allow movement of large ships from the east to the west coast of India without circumnavigating Ceylon. This is probably the only experiment carried out in open seas in the world.

The suitability of the dumping site at sea for fly ash generated by a thermal power station was confirmed by radiotracer studies using ^{46}Sc-labeled fly ash.

Groundwater Tracing

Considerable work has been done on detection of leaks in dams[9] and seepage in canals.[10] Radiotracer techniques have been used for measuring the flow in the Tapi River,[11] where discharges are up to 12 thousand m^3/sec. This work is being continued to establish the techniques for routine measurement of river flows. A laboratory is being set up for measuring low ^{3}H concentrations in water and will be used for measuring ^{3}H in both natural samples and those taken from water where tritium has been injected for groundwater studies.

Miscellaneous Tracer Work

Radiotracer techniques have been used for detecting leaks in buried pipelines[12] and in trunk telephone cables.[13] The results of these studies have allowed the industries concerned to take appropriate remedial measures.

Radioisotopes have also been used for determining retention times in chemical reactors in the rayon industry, mixing times in a sewage-purification plant, the nature of petroleum products in oil-refinery distribution lines, and wear of machine tools and other mechanical parts.

Radiation Processing

A program for the development of fiber–polymer composites utilizing cheap and abundant fibrous materials and the radiation-polymerization technique has been undertaken.[14,15] Considerable success has been achieved in making materials of improved quality, using monomers such as styrene and vinyl acetate, in the irradiation facilities at Trombay. This work is being extended for development of wood products for use in manufacture of textile bobbins, shuttles, wooden flooring, etc., using locally available low-grade wood.

Proposals for setting up an irradiator for sterilization of medical products are under consideration.

Isotopic Radiography

Isotopic radiography services are offered by the Isotope Division staff, with the staff doing the inspection, or the sources and equipment themselves are made available to the customer.

Radiography training is also provided to industrial personnel.

CONCLUSION

The radioisotope program in India has made considerable progress during the last 12 years. The availability of new and improved facilities for production and application of radioisotopes will help to achieve the objectives of the peaceful uses of atomic energy for the development of technology and economic growth.

ACKNOWLEDGMENT

This paper reviews the work carried out at the Isotope Division, Bhabha Atomic Research Centre, Trombay, and includes contributions from a large number of our colleagues, which we gratefully acknowledge. (MG)

REFERENCES

References marked with an asterisk have not been verified by the editors; for further explanation, see the Foreword.

1. V. K. Iya, An Isotope Production Programme in India, in *Programming and Utilization of Research Reactors*, Symposium Proceedings, Vienna, 1961, Vol. 3, pp. 417-424, Academic Press Inc., New York, 1962.
2. V. K. Iya, Isotope Production at Trombay, at IAEA Study Group Meeting on the Utilisation of Research Reactors, Bombay, 1964.
3. *V. K. Iya, Radioisotope Production Programme in India, at IAEA Study Group Meeting on Isotope Production, Lucas Heights, Australia, 1968.
4. *V. K. Iya, Synthesis of Labelled Compounds, at IAEA Study Group Meeting on Isotope Production, Lucas Heights, Australia, 1968.
5. N. G. S. Gopal and V. K. Iya, Quality Control of Radiopharmaceuticals, *J. Sci. Ind. Res.*, **26**(4): 153-158 (1967) (*Nucl. Sci. Abstr.*, **22**: 23586).
6. P. R. Roy, R. G. Deshpande, and T. S. Laxminarayan, Preparation of a Plutonium–Beryllium Alloy Neutron Source, Indian Report BARC/294, 1967 (*Nucl. Sci. Abstr.*, **22**: 5707).
7. Radiotracer Studies Conducted in India to Follow Movement of Materials on Sea and River Bed, Indian Report BARC/I-33, 1969.
8. V. K. Iya, A. C. Eapen, and Kirti, Radiotracer Experiment at the Sethusamudram Canal Site, Indian Report AEET/ISOTOPE/6. 1966.
9. V. K. Iya, K. Krishnamurthy, K. S. Aggarwal, and R. Rajagopalan, A Radiotracer Study on Flow Path of Seepage in the Coffer Dams at Srisailam, *Int. J. Appl. Radiat. Isotop.*, **18**: 349-357 (1967).
10. S. M. Rao, K. S. Aggarwal, B. L. Campbell, and S. V. Choughule, Radiotracer Experiments for Ganga Canal Seepage Measurements, Phase II, Indian Report BARC/ISOTOPE/14, 1967.
11. A. C. Eapen and S. M. Rao, Open Channel Flow Measurements in India Using Radioisotopes, *Indian Standards Institute Bulletin*, **19**(8): 358-361 (1967).
12. K. Krishnamurthy, S. M. Rao, R. Rajagopalan, and Kirti, Radiotracer Experiments To Locate Leaks in a Buried Water Main, Indian Report BARC/ISOTOPE/19, 1968.
13. A. C. Eapen and S. M. Rao, Leak Detection on Telephone Trunk Cables Using $CH_3{}^{82}Br$ Gas, Indian Report BARC/ISOTOPE/11, 1966.
14. V. K. Iya, Status and Technology of Polymer-Containing Fibrous Materials in India, in *Impregnated Fibrous Materials*, Study Group Report, Bangkok, 1967, International Atomic Energy Agency, Vienna, 1968 (STI/PUB/209).
15. *T. M. Jayaraman and R. C. Thacker, Studies on the Preparation of Fibre–Polymer Composites from Styrene Systems by Radiation Polymerisation, at IAEA Symposium on Utilisation of Large Radiation Sources and Accelerators in Industrial Processing, Munich, Aug. 18–22, 1969.

XUM

Portable ^{170}Tm Gamma-Radiography Unit for Light Metals*

A portable unit—weighing a few kilograms—for gamma radiography of light metals has been designed and constructed in Brazil. The gamma-radiation source is 4 or 8 Ci of ^{170}Tm (or 2 to 3 Ci of ^{192}Ir), and the cost of the unit is about a tenth that of a 100-kV X-ray machine, which gives comparable radiographs. The 4-Ci ^{170}Tm source is located at the end of a cylindrical tube inserted in an almost spherical shield, which can be rotated 180° between the exposure and storage positions (Fig. 1). In the higher-strength unit, the source is located inside the cylinder (Fig. 2). The unit performed satisfactorily in tests on aluminum welds.

(MG)

*Abstract of paper (in Portuguese) by A. C. Penteado Filho and W. Sanchez in *Metalurgia* (São Paulo), **24**: 425-432 (1968).

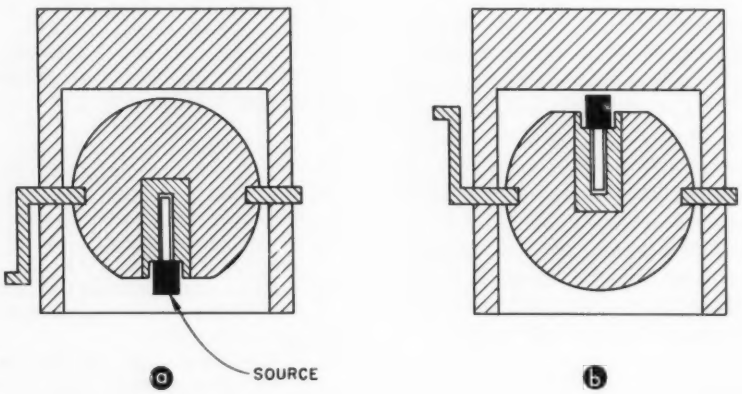

Fig. 1 Portable 4-Ci ^{170}Tm gamma-radiography unit in (*a*) exposure and (*b*) storage positions.

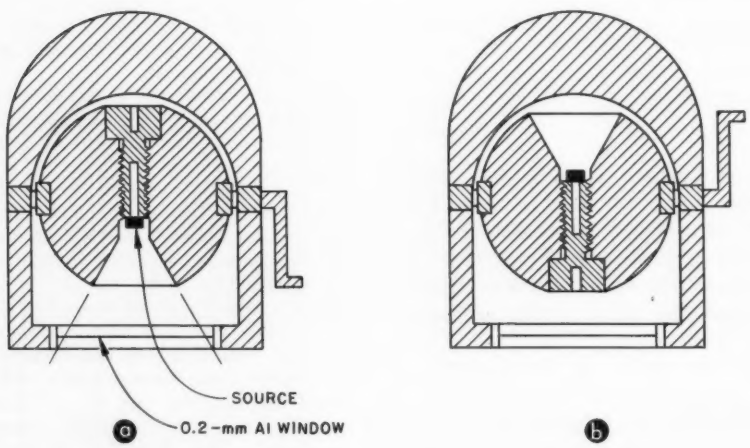

Fig. 2 Portable 8-Ci ^{170}Tm (or 2- to 3-Ci ^{192}Ir) gamma-radiography unit in (*a*) exposure and (*b*) storage positions.

Sealed Radioisotopic Sources at Nuclex 69

Several facilities and units using sealed radioisotopic sources were demonstrated at the Swiss Nuclear World Fair, Nuclex 69, in Basel, Switzerland, Oct. 6–11, 1969.

A new French power source,[1] the Gisete 6 (Fig. 1), uses $^{147}Pm_2O_3$ as fuel. The main specifications are

Overall size	170 mm diameter, 400 mm high, 10 kg
Dose rate at contact	<200 mR/hr
Active pellet size	21.3 mm diameter, 17.6 mm height
Capsule material	Hastelloy alloy C (2 sheaths)
Encapsulated source size	27 mm diameter 27 mm height
Thermal power*	5.85 W
Operating ΔT	160°C
Thermoelement material	Bi_2Te_3

	Before converter	After converter
Open-circuit voltage	1.53 V	
Output voltage at nominal charge	0.855 V	24 V
Nominal power	216 mW	180 mW
Internal resistance	3.40 [ohms]	
Overall efficiency	3.70%	3.10%
Output impedance		2900 [ohms]

*With the same source geometry, a thermal power of 8.6 W can be obtained.

Two Russian[2] power sources, both fueled with ^{90}Sr, were demonstrated—the Beta-C and the Efer.* Both convert radioactive decay energy directly to electric power and have removable heat-discharge systems. They are designed to supply electric power to automatic terrestrial units in remote or difficult-access areas with an ambient temperature ranging from +60 to −60°C. They have a service life of 10 years. Their specifications include

	Beta-C	Efer
Electric power	10 W	30 W
Rated voltage	6 V	32/12 V
Electric power capacity	880 kWh	1000 kWh
Weight	150 kg	250 kg
Weight including shipping container	500 kg	1000 kg

*Editor's Note: on handouts at the exhibit ЭФИР was transliterated as "Efer."

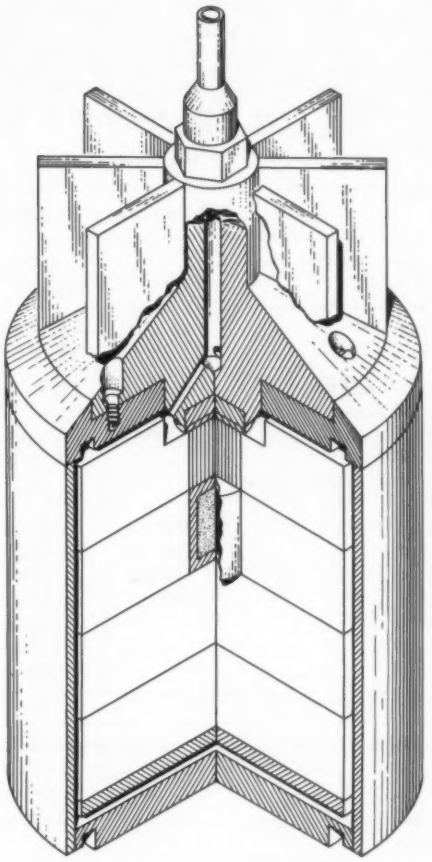

Fig. 1 Gisete 6, ^{147}Pm-fueled power (0.18 W) generator.

Other features noted on handouts at the exhibit are the unit in the Beta-C for accumulating electric energy which makes it possible to maintain the power within 1 or 2 kW under pulsed-operation conditions. In the Efer, conversion to electric power is effected by a semiconductor thermoelement unit. The system sustains a short circuit for unlimited time. Operational reliability is achieved by making the source independent of temperature and by simplicity of design; safety is ensured by a biological shield.

Two Russian ^{137}Cs irradiation facilities for presowing irradiation of seed were also demonstrated. One, the Hydroponica,* is a miniature facility, which

* ГИДРОПОНИКА.

XUM

can be installed in any building—on a stand or directly on the floor—and requires no special maintenance. The irradiator consists of 16 radioisotopic sources located at the circumferences of two parallel-plane, concentric circles. The other, the Koltos, is transportable on a ZIL-131 truck, and the operating staff is three men, including the driver. It requires no additional shielding. Some ot the parameters of the two facilities are

	Hydroponica	Koltos
Throughput capacity		1 metric ton/hr
Source activity	1.0 Ci	1300 Ci
Irradiation chamber volume	340 ml	
Dose rate	56 R/hr	800 R/min
Efficiency		22.6%
Dose-field uniformity	±20%	±20%
Maximum dose rate		
At facility surface	0.3 mR/hr	2.8 mR/hr
At truck body surface		0.25 mR/hr
Size of seeds irradiated (max.)		
Facility		
length	480 mm	7100 mm
width	500 mm	2460 mm
height	650 mm	3150 mm
weight	400 kg	3500 kg (including ancillary equipment)

Other specifications given on the Koltos handout are

Grain flow regulation range (height)	0–40 mm
Grain material characteristics	
Average bulk weight	0.7 g/ml
Average humidity	10–12%
Facility power supply (from plant in truck body or from a separate a-c supply)	220 or 380 V
Power input	1 kW

The weight and dimensions of the truck and facility are such that the loaded vehicle can travel any motor road. Speeds are quoted as maximum of 50 km/hr on a highway, averaging 35 km/hr, and an average speed of 20 km/hr on a cart road. Ranges of travel are 500 km on a highway and 400 km on a cart road. The maximum lateral list is 15°.

A Russian portable backscattering thickness gage that was demonstrated contains 0.1 mCi of ^{60}Co and is designed for unilateral thickness measurements of sheet materials, tubes, and vessel walls. The transistorized device consists of a control panel and a gage. Although calibrated with soft steel, it may also be used with other metals, the measurement accuracy being provided by calibration of the device according to the customer's standards and the operating conditions of the gage. Some of the specifications are

Measuring range	
Sheet thickness	0.5–16 mm
Tube wall thickness	2–16 mm
Error	
From 0.5 to 2 mm	0.3 mm, max.
From 2 to >16 mm	±10% of measured value
Temperature range	−10 to +40°C
Relative humidity	Up to 98% at temperatures up to 25°C

One battery set is sufficient for 50 hr of continuous operation (six "Saturn" type 1,6 FMZ-U-3,2 batteries). Less than 1 min is needed for one measurement. (MG)

REFERENCES

1. For an earlier report on French power sources, see D. N. Hess, A Summary of Recent Work by the French in the Development of Isotopic Thermoelectric Generators, *Isotop. Radiat. Technol.*, 7(1): 99-107 (Fall 1969).

2. For earlier reports on Russian sources, see Martha Gerrard, Recent Isotopic Source Developments in the USSR, *Isotop. Radiat. Technol.*, 6(2): 205-208 (Winter 1968–1969); ^{60}Co Irradiation Facility of the L. Ya. Karpov Physical Chemistry Research Institute, *Isotop. Radiat. Technol.*, 5(1): 59-60 (Fall 1967); Russian Irradiation Facility Development, *Isotop. Radiat. Technol.*, 6(1): 114-115 (Fall 1968); also p. 461 of this issue.

Gamma Irradiator Using Spent Fuel Elements*

A gamma-irradiation facility using spent fuel elements from a nuclear reactor (the VVR-M) as the radiation source has been installed at the Institute of Physics of the Academy of Sciences of the Ukrainian SSR. Tests

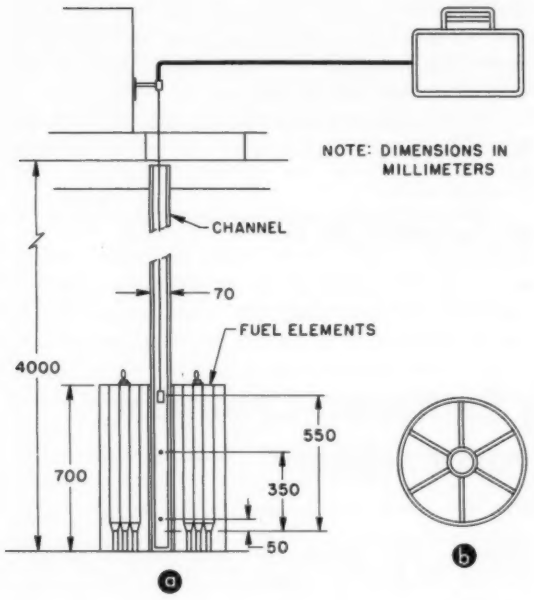

Fig. 1 Gamma irradiator using spent fuel elements. (*a*) Vertical and (*b*) horizontal sections.

with a water phantom have indicated that the dose-field intensity of the irradiator is equal to that of a ^{60}Co facility and that the useful irradiation volume is greater.

The cylindrical irradiator has a central channel surrounded by a six-compartment shell (Fig. 1), each compartment containing three fuel elements. The fuel elements used had been in the reactor for 2 years. The average energy of the radiation at the midpoint of the fuel-element length was approximately 0.74 MeV and at the ends, 0.9 ± 0.1 MeV. The mid-point value decreased to ~0.65 MeV in a month. Measured dose rates along the irradiator axis in samples of lead, copper, graphite, and a polymeric material ($C_aH_bN_cO_d$) were

Material	Sample Weight, g	Size, mm	(1)	(2)	D_m/D_{Pb}	(3)
			50	850	1.0	0.9
Pb	3.73		350	1950	1.0	0.74
		$\phi = 5$	550	1000	1.0	0.85
		$h = 16$	50	420	0.495	0.735
Cu	3.631		350	960	0.49	0.73
			550	575	0.575	0.85
			50	565	0.66	0.93
C	0.732		350	1050	0.54	0.75
		$\phi = 6$	550	580	0.580	0.82
		$h = 15$	50	610	0.72	1.0
Polymer	0.678		350	1050	0.54	0.735
			550	655	0.655	0.88

(1) Distance from bottom, mm.
(2) Dose rate (D_m), rads/sec.
(3) Average energy, MeV.

The decrease in dose rate over a month's time was

Date	Dose rate, rads/sec Pb	Cu	Polymer
Feb. 1	2020	985	1075
Feb. 3	1830	940	960
Feb. 7	1720	860	820
Feb. 17	1290	630	590
Feb. 28	995	455	450
March 1	990	420	440

(MG)

*Summary of article from *Atomnaya Energiya*, **25**: 451-453 (1968) (in Russian), by V. S. Karasev, S. S. Ogorodnik, Yu. L. Tsoglin, Yu. A. Tsybl'ko, I. M. Gorbach, V. A. Semenov, I. A. Duritskii, and V. D. Popov.

XUM

10 Years of Radioisotopes in East Germany*

Since Nov. 6, 1958, the Radioisotope Center at Rossendorf, near Dresden, has been making radioactive materials for use in medicine, scientific research, and industry. The radioisotopes are made in the 5-W WWR-S reactor, with a flux of 10^{14} n/(cm^2)(sec), and the Soviet Type U-120 28-MeV cyclotron.

In the first 10 years of its existence, the Rossendorf Center made more than 30 thousand deliveries of radioactive preparations, almost half of which were for export. The total radioactivity represented by this material was 1.3 kCi, and the monetary value, 8 million marks.† The Center today is a leading supplier of radioactive materials in Europe. It offers more than 5 hundred items, including radioisotope-labeled organic compounds and pharmaceuticals and implantable radioisotope sources for therapeutic uses. Neutron activation services are provided, as well as labeling of compounds supplied by customers. Materials are shipped out daily by rail or by air.

The Rossendorf building was constructed with the help of Soviet technical experts. (MG)

*Based on brochure, *10 Jahre Radioaktive Präparate, Rossendorf (ROTOP)*, ISOCOMMERZ GmbH, Lindenberger Weg 70, 1115 Berlin-Buch.

†One East German mark is equivalent to about 45 cents at the official exchange rate.

Economics of Industrial Radiation-Chemical Sulfonation of Hydrocarbons*

A radiation-chemical plant for sulfonation of hydrocarbons was started up in 1967 in the USSR, the first of its kind in that country. The process uses radiation to convert a mixture of hydrocarbons ("synthine") to sulfonate at a net rate of 360 kg/hr, or 2600 metric tons/year with continuous operation for 300 days/year. The capacity of a photochemical reactor in the process, as formerly carried out with a mercury-vapor lamp, is about 40% of that of the radiation-chemical reactor.

Use of the radiation-chemical method, with a ^{60}Co source, resulted in a total annual saving of 200 thousand rubles† in the production of 6500 metric tons of sulfonate. The radiation-chemical reactor (model RS-2.5), including the radiation source, cost 80 thousand rubles. Some 70 thousand rubles worth of ancillary equipment was eliminated, and the overall saving in space represented a saving of 140 thousand rubles.

Hydrolysis of the sulfonyl chloride reagent is 1.5- to 2-fold greater in the radiation-chemical process than in the photochemical, and somewhat more water is used. However, less oleum and other materials, coolant, and electric power are consumed. Annual economies resulting from these savings were, in thousands of rubles, 28.5 for materials, 4.5 for electric power, and 6 for coolant. Deducting 6.4 thousand rubles for the increase in water needs gives a net saving of 32.6 thousand rubles per year.

Unit costs for the product are quoted as 19 rubles per metric ton for the capital investment and 822 rubles per metric ton for such items as materials, energy, wages, and overall amortization. Amortization of the radiation source is only 0.3% of the net unit cost of 822 rubles.

(MG)

*Based on V. B. Osipov, V. P. Guteev, R. V. Dzhagatspanyan, A. I. Gershenovich, and S. V. Golubkov, Technico–Economic Aspects of Radiochemical Production of Sulfonates (in Russian), *At. Energ. (USSR)*, **25**: 320-321 (1968); and R. V. Dzhagatspanyan, L. M. Yakimenko, V. I. Sinitsyn, Yu. G. Lyaskin, V. I. Zetkin, and B. Ya. Libman, Radiation-Chemical Sulfochlorination of Kerosene and Synthine (in Russian), *Khim. Prom.*, No. 4, pp. 7-11 (247-251), 1965.

†One ruble is equivalent to $1.11 at the official exchange rate.

Quick Balance of Mercury in Electrolytic Cells by Using Radioactive Mercury

N. Pasi and R. Gvion have reported[1] in the *Israel Journal of Chemistry* a practical method for determining the balance of mercury in electrolytic plants producing chlorine and caustic soda by the mercury-cathode method. The old method of stopping the plant (where there are tens of tons of mercury in circulation) to check for mercury losses (leakage, evaporation, etc.) by draining and weighing all of the mercury is expensive and time consuming. A new method that uses radioactive mercury and is much faster was developed.

Three grams of mercury was irradiated in the reactor and then diluted with 60 g of unirradiated mercury. A quantity of this tracer was introduced into the flowing cathode of one of the electrolysis batteries; the dilution factor was about 1 to 10 thousand. After 3 hr the mercury was homogeneous. The activity of the tracer at the time of introduction was about 200 mCi of ^{197}Hg (65-hr half-life) and 12 mCi of ^{203}Hg (47-day half-life). All the samples taken from a battery, together with reference samples of the original tracer, were counted within a half hour to avoid the decay error in counting the short-lived isotope. The authors state that the error due to decay during this period is less than 1%. Each sample gave at least 20 thousand counts in less than 3 min for a statistical error of less than 1%. Comparison of results from the counting method with those from three plant experiments using previously weighed quantities of mercury indicated an accuracy within 1.5%. Plans are now to make an overall mercury balance of the entire plant (more than 20 batteries) in a single day.

No contamination was found in the aqueous phase in contact with the cathode, and the air in the electrolysis hall showed no radioactive mercury contamination. Radiation near the battery was very low, about 4 mR/hr. The authors also stated the main component of the activity, ^{197}Hg, disappeared almost entirely in 25 days.

Gold-198 in the form of a mercury–gold amalgam, as an alternative method, was tried but was unsuccessful. (REG)

REFERENCES

1. N. Pasi and R. Gvion, Quick Balance of Mercury in Electrolytic Cells by Using Radioactive Mercury, *Israel J. Chem.*, 6: 83 (1968).

Nuclear Methods in the Aviation Industry

Amos Notea and Shlomo Shalev, *Nuclear Methods in the Aviation Industry* (in Hebrew), Technion–Israel Institute of Technology, Department of Nuclear Sciences, Haifa, Israel, Mar. 5, 1969.

A brief pamphlet, *Nuclear Methods in the Aviation Industry,* has been issued by the Nuclear Sciences Department of the Technion–Israel Institute of Technology in connection with the 11th Annual Israeli Conference on Aviation and Astronautics. The booklet describes in an elementary, nonmathematical way the principles of measurements utilizing radioisotopes as they apply to the aviation industry. Such applications as level and thickness gages, flowmeters, rotameters, distance-measuring devices, and mass detectors are covered, as are also wear and leak detectors, an air-density altimeter, and some possible uses of neutron radiography. The booklet is comparable to some of those in the USAEC's "Understanding the Atom" series. (E. G. Silver)

XUM

Coal-Ash Measurement with Two Radioisotopic Radiation Sources*

A Japanese method for determining the amount of ash in coal by the radiation-absorption technique uses two radiation sources—100 mCi of ^{241}Am (60 keV) and 30 mCi of ^{137}Cs (660 keV) (Fig. 1). The purpose of the second source is to correct for density variations, which is done from the formula

$$\mu_{(1)} = \frac{\log I_{0(1)} - \log I_{(1)}}{\log I_{0(2)} - \log I_{(2)}} \mu_{(2)}$$

where I_0 and I are the intensities of the radiation before and after it passes through the sample, μ is the mass absorption coefficient, and the subscripts (1) and (2) refer to ^{241}Am and ^{137}Cs, respectively.

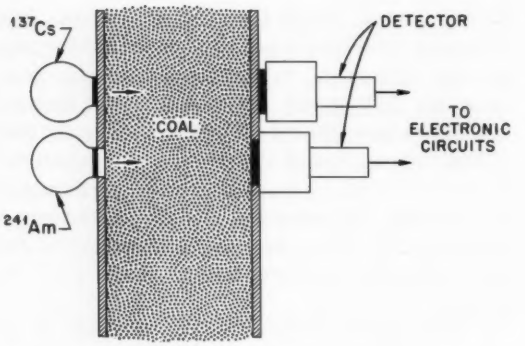

Fig. 1 Two-source system for determining ash in coal with correction for density variations.

The system was tested with raw and refined coal samples of known ash content and found to give satisfactory results. (MG)

*Summary of paper by Hiromu Fushimi in *Nucl. Eng. (Tokyo)*, 13(6): 47-51 (1967) (in Japanese) (translated into English as Japanese Report NSJ-tr-134).

Determination of Heavy-Oil Heat Value with Neutrons

The heat value of a heavy oil is usually determined directly by an adiabatic bomb method, which requires several hours per determination. A continuous, automatic gage has been developed in Japan* which contains a 10-mCi Ra–Be fast-neutron source. The neutron attenuation by the oil is a measure of the hydrogen content, and hence the fuel value, of the oil. The gage is practicable because of the absence of other high-cross-section elements, with the exception of a small amount of sulfur. A BF_3 counter is used. Tests on the method showed precisions comparable to those of the bomb method. (MG)

*K. Miyoshita, Continuous and Automatic Measurement of Heavy Oil by the Use of Neutrons (in Japanese), *Nucl. Eng. (Tokyo)*, 13(10): 52-55 (1967) (translated into English as Japanese Report NSJ-tr-152).

Editor's Note: The reader is referred to two articles on hydrogen determination in hydrocarbons by a slow-neutron transmission technique:

1. W. E. Mott and D. F. Rhodes, Hydrogen Analysis by Slow Neutron Transmission, in *Radioisotope Instruments in Industry and Geophysics*, Symposium Proceedings, Warsaw, 1965, Vol. I, pp. 347-364, International Atomic Energy Agency, Vienna, 1966 (STI/PUB/112).
2. D. F. Rhodes and W. E. Mott, Hydrogen/Density Analyzer Monitors Process Streams, *Instrum. Technol.*, 16(7): 52-55 (1969).

Survey of Radioisotope Applications in Latin America*

By G. B. Baro and J. G. Flegenheimer†

Radioisotope applications in Latin America are summarized here, with special emphasis on Argentina because the authors are naturally most familiar with that country.

Perhaps the chief reason for building the first reactor in Argentina was to train reactor physicists. However, the radiochemistry group was already a strong one, having had the advantage of several years of training with radioisotopes produced by the Buenos Aires synchrocyclotron and by a Cockcroft–Walton accelerator. Therefore, as soon as the fun was over for the reactor physicists and the reactor had been tested, it was put on a routine operating basis for producing isotopes. It is still doing this, after several redesigns.

In our experience, the first step is to get the reactor going and producing some of the ordinary relatively short-lived radioisotopes. Soon, so many people wake up to what is going on that demands for these materials grow at an exponential rate. You are then obliged to produce other isotopes to supply the needs. Demands outstrip the reactor capacity, and you soon have to import most of the isotopes. At this point, the reactor physicist comes into the picture again, very willing, of course, to design a new and larger reactor that will satisfy demands for some years. Thus you end up with a reactor designed and operated mainly for radioisotope production. Argentina is now operating its RA-3 reactor, which has a 5-MW power level with a maximum thermal flux of about 2×10^{13} n/(cm^2)(sec). A new building of about 2 thousand m^2 for radioisotope production, adjacent to the reactor, is to be completed by the end of 1969. Except for the production of high-intensity radiation sources, Argentina hopes soon to supply all its own demands and to export radioisotopes to other Latin American countries.

Besides radioisotope production, it is interesting to note the consumption of radioisotopes. In Argentina, from 1954 until 1968, the consumption of radioisotopes increased about 10-fold, from about 8 Ci to about 85 Ci (Fig. 1). We do not have the figures for other Latin American countries, but we believe that the consumption in Brazil has been roughly the same. The consumption in Latin America can therefore be estimated approximately by multiplying the figures for Argentina by 4 or 5. The long-lived radioisotopes were all imported, but the short-lived—less than a month—were at least partially produced in those Latin America countries which have reactors. We can safely assume that, with larger research reactors operating, most of the short-lived radioisotopes will be produced domestically. Perhaps consideration should be given to joining efforts among our countries and establishing large regional production centers to supply the demands of all Latin America.

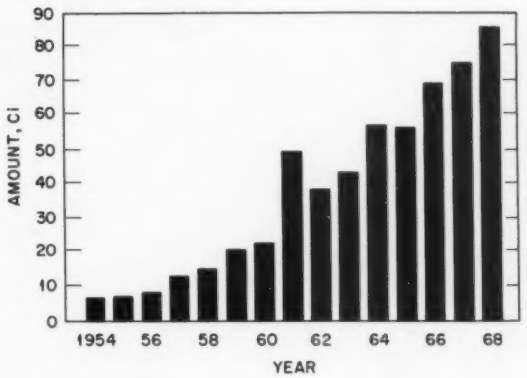

Fig. 1 Consumption of radioisotopes in Argentina exclusive of radiation sources.

USES

The main question, of course, is not how much of something is produced or consumed but what is done with it. How radioisotopes have been used in Argentina is shown in Table 1. Large variations may occur from year to year, depending on how much long-lived source material is imported for use, for example, in irradiators for medical therapy, for agricultural seed or plant treatment, or for industrial purposes. In Argentina, there are now some 35 thousand Ci of ^{60}Co, 7 thousand Ci of ^{137}Cs, and about 2 hundred Ci of several

*Shortened version of paper (in Spanish) from American Nuclear Society Topical Meeting in Puerto Rico, 1969.

†Comisión Nacional de Energía Atómica, Buenos Aires, Argentina.

XUM

other source materials—e.g., ^{226}Ra and ^{90}Sr—being used in medicine.

The Agriculture and Veterinary Department of Buenos Aires University has an 11 thousand Ci ^{60}Co gamma source for seed and plant irradiation; the Argentina Atomic Energy Commission has a 10 thousand Ci ^{60}Co and a 1 hundred Ci ^{60}Sr source for irradiating liquids and is building an irradiation plant in Ezeiza, near Buenos Aires, with a 10^6-Ci ^{60}Co source and a capacity of 1 ton/hr for 1-Mrad irradiations of material with a specific gravity of 1. It will initially operate with 250 thousand Ci of ^{60}Co.

labeled molecules prepared on request are all used in medicine or other life sciences.

The wide use in medicine has been stimulated by the Argentina Atomic Energy Commission in two ways. First, the Commission holds regular training courses on the use of radioisotopes, at which 87% of the attendants are medical doctors. Second, the Commission contributes to the support of a Nuclear Medical Center in Buenos Aires, where medical students can complete their training in nuclear medicine and medical doctors are developing radioisotopic techniques. The Commission supports work in hospitals

Table 1 Use of Radioisotope Products in Argentina: Distribution by Field

4-Year Average, 1963 to 1966

Field	Amount, % of total			No. of institutions authorized to use radioactive materials
	Radio-isotopes	Labeled compounds	Sealed sources	
Medicine	70.8	70.0	20.8	223
Industry	1.0	0.1	70.9	64
Research (mainly biology)	28.0	29.5	6.9	45
Agriculture	0.2	0.4	1.4	19

A number of irradiators in Latin America can be used by all CIEN (Comisión Interamericana de Energía Nuclear) member countries. Mexico has two Van de Graaff accelerators, 0.6 and 2 MeV, respectively; two ^{60}Co irradiators, 3700 and 2000 Ci, respectively; and one Dynamitron electron accelerator, 12 mA and 5 MeV. Ecuador has a Gammacell-200, and Brazil, in Bello Horizonte, has 4000- and 1600-Ci ^{60}Co facilities. In Central America, there is a 2400-Ci ^{60}Co source, and in Puerto Rico, an 8000-Ci ^{60}Co source and a 10,000-Ci ^{137}Cs source. Venezuela, Peru, Uruguay, and other countries also have irradiators, and Chile is obtaining a 50,000-Ci ^{60}Co source from the IAEA.

Medicine

In Argentina, radioisotopes have been used mostly in the biomedical field, as is true also for the other Latin America countries. There are 3749 authorizations for using radioactive materials in our country, and more than 90% of them are for medical and biological work. The 38 radioactive compounds now produced routinely and a similar number of special

and other institutions on biological application of radioactive materials, in many cases supplying the materials on a no-charge basis or at prices as low as 10% of catalog prices. About 150 papers on biological experiments using radioactive isotopes are published in Argentina annually.

A regional center of reference for secondary standards in dosimetry has been established at the Ezeiza Atomic Center for calibrating dosimetry equipment used in radiotherapy, for personnel training, and for advisory service to all countries in the region.

Industry

Industrial applications have developed somewhat less rapidly than medical, but they have made an impact, as shown by an inquiry carried out by the IAEA in 1963. Industrial radiography has come of age in Argentina, with more than 90 commercial firms and private specialists authorized in this field (Fig. 2). Originally, all such work was done by the Commission. With the new reactor at Ezeiza, ^{192}Ir sources will be produced.

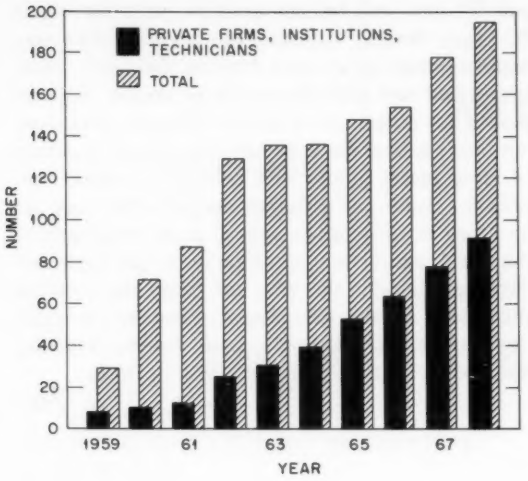

Fig. 2 Users of industrial gamma radiography in Argentina.

Radioisotopic gages for metal sheets, paper, and plastics were also developed originally by the Commission but are now produced commercially. About 50 radioisotopic gages—for measuring thickness, density, levels, and humidity—are in use in 27 factories. The Commission's policy has been to produce prototypes of the equipment and then to make the design available to private companies for manufacture. Latin American markets for such gages are still limited, but a much wider use is predicted.

Some radioisotopic techniques are carried out routinely in Argentina, but others have been developed for special purposes, e.g., for finding leaks in pipes, measuring flow rates in waterways, studying wear in motors, measuring snow thickness automatically in the Andes, studying sediment transport in the Rio de la Plata and along seashores, determining residence time in dryers, inventorying mercury in electrolytic cells, locating scrapers in large underground pipelines, and tracing materials in metallurgy.

Argentina has used delayed-neutron activation analysis for determining uranium in alloys. At the request of the Law Court, arsenic has been determined in human hair and nails and the results used in settling a lawsuit. The efficiency of motor lubricants has been measured, and recently a nondestructive method for determining arsenic and antimony in lead and tin alloys was developed. Biological samples of different origins are also being analyzed. With the RA-3 reactor and adjacent facilities now in operation, activation analysis will be used more and more in Argentina.

Other Latin American countries possessing reactors have also used modern activation-analysis methods. Brazil, for example, has determined hafnium in zirconium, sodium in demineralized reactor water and graphite, gold in minerals, and several other elements in a variety of materials. Work has also been done on activation analysis of hairs for identification. Venezuela, Colombia, and Uruguay are also interested in this field.

A very useful application of radioisotopes in Latin America is in hydrology. In Chile and in the Andean provinces of Argentina, it is very important to know in advance the amount of water that will be released to the rivers in the summer. This can be deduced from the amount of snow accumulated in the winter. Such information is useful in, for example, droughts, as occurred this year in Mendoza and San Juan. We hope that a more extensive use of radioisotopic snow thickness gages will help alleviate such problems in the future. Chile has used radioisotopes to establish the flow and direction of underground waters in Pampa del Tamarugal and in the Cantones and Salar Pintado regions in Tarapacá Province.

Latin America is also interested in applying nuclear techniques to prospecting and mining, and a recent IAEA symposium on this subject was held in Buenos Aires. Such methods have not yet been used extensively—apart from water, oil, and radioactive mineral explorations—but Latin America, with its ample natural resources, is a fertile ground for them.

Tracers have been used in agriculture and livestock studies. Colombia has used ^{32}P to study barley fertilizers; Brazil has studied fixation of carbon dioxide by nitrifying bacteria; Chile has used ^{32}P and ^{131}I to study the action of tetracycline on chicken growth. The work done at Turrialba, Costa Rica, on applying radioisotopes in tropical agriculture is known throughout America. Argentina has carried out studies on soils, lack of minerals in livestock, and labeling of insects and mice.

Large radiation sources in Latin America are used mainly for radiation therapy, but interest is growing in their application to food preservation, polymerization, chemical and biochemical effects of radiation with a practical objective, and industrial sterilization. Venezuela, Costa Rica, Peru, and some other countries have programs on sterilization of male insects to eliminate pests. Curaçao has eliminated the fruit fly in this way.

A semiindustrial irradiation plant soon to be operated in Argentina will be used mainly for industrial sterilization of pharmaceutical products and in cost studies on processes using high-intensity radiation

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sources. When completed, this plant will be one of the largest in the world.

FUTURE

Latin America is mainly an exporter of raw materials, but this situation is changing. Rapid industrialization and population increases will call for greater power output, and nuclear power is an obvious solution to some of the problems. Argentina will have its first power reactor [316 MW(e)] operating in 1972, and advances in nuclear know-how will be accompanied by an expansion of radioisotope applications. With the production of fissile material in power reactors, the transuranium by-products—e.g., ^{237}Np and ^{241}Am—will become available, probably within 10 years. Nuclear batteries will certainly find very useful applications in Latin America, and costly installations and new technologies will be needed. Regional centers for nuclear work would be a great advantage, not only for favorable economics but also for exchange of ideas among groups with common interests. Pressures for results and getting things published is not yet so strong in Latin America as in some other parts of the world, and there is still time for sitting down and thinking originally and even for trying out methods which have a rather dim chance of success but which may provide useful shortcuts. Latin America is moving fast, and we are most optimistic about its future.

(MG)

Section

V

Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

ORNL STABLE-ISOTOPE INVENTORY AUGMENTED

Recent additions to the Oak Ridge National Laboratory (ORNL) enriched-stable-isotope saleable inventory include the following:

Isotope	Purity, %	Amount available, g
^{10}B	99.9	7.9
^{11}B	99.8	8.9
^{33}S	85.9	1.1
^{34}S	90.0	2.5
^{50}Cr	96.8	11.1
^{54}Cr	95.4	4.5
^{54}Fe	92.2	17.9
^{62}Ni	98.8	96.1
^{77}Se	66.6	4.9
^{77}Se	61.1	5.7
^{98}Mo	97.3	253.4
^{120}Te	26.7	0.1
^{130}Ba	58.5	0.08
^{130}Ba	48.8	0.2
^{130}Ba	37.0	0.3
^{132}Ba	30.8	1.2
^{132}Ba	21.6	1.5
^{134}Ba	59.4	2.2
^{135}Ba	78.8	3.8
^{135}Ba	56.5	8.2
^{137}Ba	64.0	12.7
^{158}Dy	20.8	0.7
^{164}Dy	98.4	17.7
^{167}Er	91.5	68.1
^{168}Er	95.5	40.0
^{170}Er	96.9	40.0
^{196}Hg	48.2	3.0
^{196}Hg	27.0	0.4
^{208}Pb	99.9+	1.6
^{208}Pb	99.8–	1.6

The AEC has recently announced new prices for electromagnetically enriched ^{151}Sm, a beta-emitting radioisotope with a half-life of ~87 years:

6–13% enrichment	$71.00 per gram (minimum sale $100.00)
75–90%	$23.00 per milligram
>90%	$36.00 per milligram

In addition to the regular inventory, ORNL maintains a rather large research materials collection; for further information regarding availability from this inventory, write Isotopes Sales Department, Oak Ridge National Laboratory, P. O. Box X, Bldg. 3037, Oak Ridge, Tenn. 37830. (PSB)

POLONIUM-210

The AEC has raised the price of ^{210}Po to $80.00 per curie in order to recover production costs, which have increased following a drop in demand. The decrease in demand resulted from the recent cancellation of the SNAP-29 program on development of a radioisotope thermoelectric generator fueled with ^{210}Po for space flight missions. Cancellation was the result of budgetary and program priority considerations, not technical feasibility. The previous price for ^{210}Po metal was $15.00 per curie for the most commonly ordered quantities, i.e., a few hundred curies. Commercial demand totaled about 4500 Ci in 1968, but the SNAP-29 program used many times that amount.

Polonium-210 production and commercial distribution are conducted by Mound Laboratory at Miamisburg, Ohio, which is operated for AEC by Monsanto Research Corporation.

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RADIANT ENERGY CORP. OFFERS CONSULTING SERVICES

A new radiation consulting firm has been set up to provide consulting services for private companies and government agencies that are using, or wish to use, irradiation as an industrial process. The firm, Radiant Energy Corp., is located at 1701 N. Fort Meyer Drive, Suite 603, Arlington, Va., with John Masefield as president (formerly with Atomic Energy of Canada) and Carl B. Wootten (formerly with Isotopes). The firm also has a Canadian subsidiary, Masefield Irradiation Consultants, Ltd., at Montreal.

McMASTER UNIVERSITY OFFERS NUCLEAR ENGINEERING PROGRAM

In response to the increasing importance of nuclear power in Canada and the needs of industry for professional personnel specifically qualified in nuclear engineering, McMaster University, Hamilton, Ont., will take advantage of its 2-MW(e) pool-type research reactor to provide an appropriate program of teaching and research in this field. Under the guidance of Dr. Archie A. Harms, who holds a Ph.D. degree in nuclear engineering from the University of Washington, the program calls for an elective in nuclear engineering for fourth year undergraduate students in science and engineering; an area of specialization in nuclear reactor physics and nuclear engineering systems for graduate students; and an opportunity for professional personnel in the nuclear industry to update and extend their academic–professional qualifications. There will be close liaison with selected representatives from the nuclear industry to ensure a well-balanced program closely attuned to the academic and industrial objectives of current importance.

BRITISH COURSES IN RADIOISOTOPE WORK

A 2-week course, "Introduction to Radioisotopes," was offered in the fall of 1969 at the Post-Graduate Education Centre, A.E.R.E., Harwell, England, and is scheduled again for spring and fall in 1970. The course is a general theoretical and practical introduction to the principal techniques used in tracer applications of radioisotopes in research and technology, with reference to the use of higher levels of radiation. It is not directed toward any particular discipline or application but is designed to be followed by a specialist course or to stand alone as a general introduction. The work may include gamma radiography, measurement of neutrons, autoradiography, or additional radiochemical work.

^{124}Sb–Be NEUTRON SOURCE

Two ^{124}Sb–Be neutron sources—the Neutroncell 200 and the Neutronpool 500—are being offered by AECL Commercial Products. The sources, which provide fluxes of 2×10^8 and 6×10^8 n/(cm^2)(sec), respectively, consist of a high-specific-activity mass of ^{124}Sb surrounded by a large amount of beryllium (Fig. 1). The units can be used to produce a number of short-lived tracers—e.g., ^{99}Mo, ^{64}Cu, and ^{56}Mn—and more than 90 radioisotopes with half-lives of less than 200 min can be made with specific activities of $>0.1\mu$Ci/g. Other uses of the neutron sources include neutron activation analysis and process control. In terms of dollar volume, ^{124}Sb should prove to be very important in Canada.

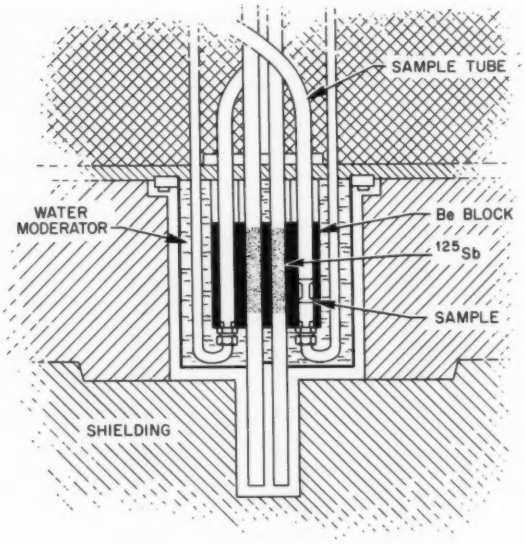

Fig. 1 Canadian neutron source.

RADIATION RESOURCES, INC., OFFERS NEW GAMMA IRRADIATOR

Radiation Resources, Inc. (24 Kulick Road, Fairfield, N. J. 07006), is offering its first irradiation equipment product, the "Scholastic Radatron," an irradiator for experimental use in unrestricted areas of schools, hospitals, and industry. It has four chambers,

three of which have a total volume of 600 ml and the fourth, 1000 ml. The radiation rate in the chambers varies from 3000 to 37,000 R/hr. One model contains 400 Ci of ^{137}Cs.

ORNL SUPPLIES GALLIUM-67

The USAEC has granted approval to ORNL's Isotopes Development Center to produce and distribute experimental quantities of ^{67}Ga for development and evaluation. The radionuclide is supplied as a chemical-grade chloride solution of carrier-free ^{67}Ga in dilute HCl. An interim price of $10/mCi, plus the standard $25 handling charge per shipment, FOB Oak Ridge, Tenn., has been established. For further information, write Isotopes Development Department, Isotopes Development Center, Bldg. 3047, P. O. Box X, Oak Ridge, Tenn. 37830.

AEC Activities

^{252}Cf NEWS LETTER

A series of progress reports on the ^{252}Cf market evaluation program being conducted by the U. S. Atomic Energy Commission is to be issued. These reports augment the brochure *Californium-252, Its Use and Market Potential.* Each issue of *Californium-252 Progress* will consist of two main sections:

News: Reports on participants in the program, cost and availability of ^{252}Cf, market projections, announcements of meetings and publications, and other topics related to the development of ^{252}Cf applications, which are expected to lead, eventually, to a large market for the nuclide.

Technology Development: Reviews of applications technology developed under the market evaluation program, californium production technology, and other technical accomplishments related to ^{252}Cf utilization.

Copies of the brochure and progress reports may be obtained by writing to Manager, Savannah River Operations Office, U. S. Atomic Energy Commission, P. O. Box A, Aiken, S. C. 29801. The contents of the first issue (October 1969) were

News

Texaco and Schlumberger evaluating ^{252}Cf in well logging for oil and gas reserves
Considerable response received to market evaluation program
^{252}Cf can be produced with present technology
Biomedical Advisory Panel coordinates evaluations of biomedical applications
Procedures given for applying for license to handle sources

Applications Development

Radiotherapy
Neutron radiography
Natural resources exploration
Process control

General

Shielding evaluations
Soil moisture and density measurements
In-line activation analysis

Bibliography

AEC SIGNS MORE LOAN AGREEMENTS FOR USE OF ^{252}Cf

Since the issue of *Californium-252 Progress* (see the item above), two commercial firms and a research institute have recently signed contracts with the AEC for the loan of ^{252}Cf for practical application studies.

The Fort Worth Division of General Dynamics Corporation, Fort Worth Tex., will use a 1000-μg source in developing neutron radiographic methods for examining aircraft parts and systems. GeoSensors Inc. of Dallas, Tex., a subsidiary of Gulf Resources and Chemical Corporation, Houston, will use two sources of 50 and 500 μg, respectively, in neutron activation as applied to mineral exploration and rock and mineral analysis. The Columbia Scientific Research Institute of Austin, Tex., will use five sources of 80 μg each to detect impurities in ores and bulk materials using the neutron activation technique as the material passes by the sources.

Loan agreements have been signed previously with Republic Steel Corporation, Cleveland, Ohio, for 100- and 1000-μg sources for process control studies. Texaco Inc., Bellaire, Tex., is doing mineral exploration studies with 61- and 703-μg sources, and Schlumberger Technology Corporation, Ridgefield, Conn., is doing similar studies with sources of 48 and 430 μg. Loan agreements have also been signed with five other organizations, including two medical institutions which are studying its applicability in cancer therapy.

XUM

The sources will be fabricated at the AEC's Savannah River Plant near Aiken, S. C., operated for the Commission by the E. I. du Pont de Nemours & Company.

AEC ORDERS 10 SNAP-19'S FROM ISOTOPES, DIVISION OF TELEDYNE

The AEC is ordering 10 modified SNAP-19 ^{238}Pu-fueled generators and backup systems from Isotopes for use by NASA in unmanned Jupiter and Mars space missions. The generators are rated at 30 W each. Four of the generators will be used in each of two Pioneer missions, scheduled to fly past Jupiter in 1972 and 1973, respectively, and two in the Viking project, scheduled for a soft landing on Mars in 1973. The modification over the SNAP-19 generator in the Nimbus-B-2 weather satellite is a change in the plutonium fuel form and a new thermoelectric system that uses germanium telluride.

RADIOISOTOPE-POWERED SELF-ILLUMINATING TRAFFIC SIGN

The Arizona Highway Department is testing a self-illuminating lane-control sign in Phoenix which was designed and built by American Atomics Corp. The sign contains a phosphor activated by ^{85}Kr, which was enriched at ORNL. The sign requires no electrical connections, is easily visible under ordinary night conditions from 500 ft, and is designed to last at least 10 years (see Fig. 1).

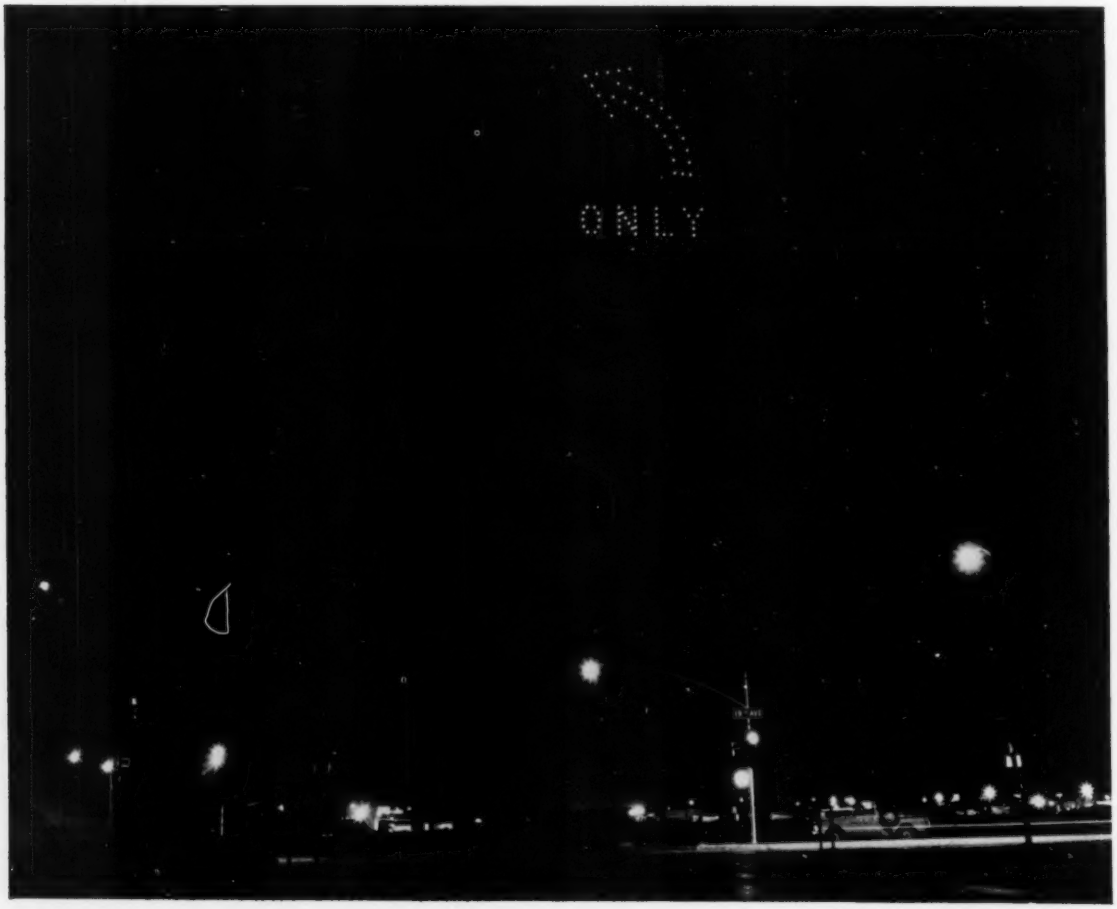

Fig. 1 American Atomics Corp. ^{85}Kr-illuminated highway sign.

AEC APPROVES AGREEMENT WITH GEORGIA

The AEC has approved a proposal by the State of Georgia for an agreement under which Georgia will assume part of AEC's regulatory authority over the use of radioactive materials in that State. This is the twenty-second agreement state. The transfer of regulatory responsibility includes licensing, rule making, and enforcement in the uses of radioisotopes, the source materials uranium and thorium, and small amounts of fissionable materials. There are about 260 AEC licensees in Georgia for the use of radioactive materials. The Department of Public Health will administer the State's radiation control program, including the use of radioactive material, X-ray machines, and other sources of radiation not regulated by AEC. The Commission has found that the radiation control program proposed by Georgia is compatible with that of the AEC and is adequate to protect public health and safety.

SIX SCIENTISTS NAMED TO AEC ADVISORY GROUP

Six scientists from the fields of education and industry have accepted appointment to the Atomic Energy Commission's Advisory Committee on Isotopes and Radiation Development. This committee meets about every 6 months to guide the Commission's isotope development program.

John W. Landis, San Diego, Calif., Group Vice President, Engineering and Manufacturing, Gulf General Atomic, Inc., heads the 12-member committee. New members are Dr. Merrill A. Bender, Department of Nuclear Medicine, Roswell Park Memorial Institute, Buffalo, N. Y.; Dr. Merril Eisenbud, Professor of Environmental Medicine, New York University Medical Center, and formerly the Administrator of the Environmental Protection Administration of the City of New York; Dr. Ira Lon Morgan, Director of the Center for Nuclear Studies, the University of Texas at Austin; Dr. Lyle E. Packard, President of Packard Instrument Co., Inc., Downers Grove, Ill.; Dr. A. J. Restaino, Manager, Radiation and Plastics Development, Atlas Chemical Industries, Inc., Wilmington, Del.; and Dr. Seymour Rothchild, President, New England Nuclear Corporation, Boston, Mass. Other members of the committee are: Dr. Nathaniel F. Barr, Division of Biology and Medicine, AEC; Dr. Milton Burton, Director of Radiation Laboratory, University of Notre Dame; Dr. Bernard A. Fries, Chevron Research Co., Richmond, Calif.; Dr. David E. Harmer, Radiochemistry Research Laboratory, Dow Chemical Co., Midland, Mich.; and Robert Kettner, President, Nuclear Assurance Corp., Atlanta, Ga.

PAUL C. AEBERSOLD MEMORIAL LECTURE

Dr. Glenn T. Seaborg gave the memorial lecture, "The Expanding Role of the Atom in the Humanities," on Dec. 2, 1969, at the American Nuclear Society's meeting in San Francisco. Dr. Aebersold, who died in 1967 [*Isotopes and Radiation Technology*, **5**(1): iv (Fall 1967)], was for many years Director of the AEC's Division of Isotopes Development.

General

NUCLEAR MEDICINE RECOGNIZED AS A SPECIALTY

At a Fall, 1969, meeting, the American Medical Association's (AMA) Liaison Committee for Medical Specialties approved the concept of nuclear medicine as a special field for physicians. The AMA was studying the mechanism for setting up a nuclear medicine board and was expected to create such a board at its January 1970 meeting. The Association's action will provide a means for establishing and supervising resident training programs and the mechanism for ensuring competence of physicians. The American Society of Nuclear Medicine has been urging this step for some time.

DEVELOPMENTS IN RADIOISOTOPIC DIAGNOSES

Developments in certain nuclear medicine techniques, which can be performed entirely in the laboratory with no administration of radioactive materials to patients, have led to new diagnostic methods. This was reported at a meeting, in September 1969,

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titled "In Vitro Procedures with Radioisotopes in Clinical Medicine and Research," called by the International Atomic Energy Agency (IAEA) and the World Health Organization (WHO). More than 200 specialists in medical work and in the use of radioisotopes—from nearly 30 countries and three international organizations—attended the meeting.

The new procedures are simple, highly sensitive, and low in cost. They can, in most cases, be performed in small medical radioisotope laboratories with relatively simple apparatus, and they can be used where facilities are limited or where field studies must be made. Much work remains to be done, however, especially on standardization, before these methods can be used routinely.

Topics discussed at the meeting include labeling of cells with radioactive materials for the diagnosis of cancer and other diseases, as well as for medical research; immunological studies to determine quickly whether mass immunization against an infection is necessary or not; investigations of thyroid diseases where much work may be saved by blood studies instead of administering radioactive iodine to patients; and ways of measuring hormones, including insulin and growth hormones.

At another meeting held at the IAEA in September, in a 4-day session the Panel on Isotope Procedures in Diagnostic Hematology (a subsection of the International Committee for Standardization in Hematology) reached agreement on certain methods of diagnosing blood diseases by means of radioisotopes. Representatives of the IAEA and the WHO also attended the meeting, bringing the number of participants to 13, drawn from 9 countries. The extent of the experts' agreement marked a definite step toward the formulation of worldwide standards to guide physicians in the use of radioisotopes in hematology.

For determining the life-span of red blood cells, important in diagnosing anemia, for example, a sample of the patient's blood is labeled with ^{51}Cr and reinjected. The labeled cells can subsequently be detected in the blood throughout their life-span—about 115 days in normal persons. A compound of phosphorus and fluorine, known as DFP, labeled with ^{32}P, ^{14}C, or ^{3}H, can be used similarly. The Panel reached agreement on standard techniques for labeling red cells with ^{51}Cr, on methods of studying red-cell survival with both ^{51}Cr and labeled DFP, and on methods of presenting and interpreting the data. Draft recommendations concerning these techniques and methods were drawn up and, if approved at the 13th International Congress of hematology in Munich, in August 1970, will be published in the world's leading hematological journals.

SUCCESSES IN STERILE INSECT FRUIT PEST CONTROL

Results of experiments in which more than a billion Mediterranean fruit flies were reared, irradiated, and released among wild populations have shown that the radiation sterilization technique of controlling this pest is effective. The work, reported at an IAEA/FAO-sponsored meeting in Vienna in September 1969, was sponsored by international and national organizations, including the USAEC. The experiments were carried out in Central America, Italy, and Spain and involved examination of tons of fruit—oranges, apricots, and peaches—as well as of trapped insects.

RADIATION-INDUCED MUTANTS

Contributions to the Green Revolution by plant breeders using radiation techniques to improve crops throughout the world are described in Vol. 11, No. 5, of the International Atomic Energy Agency's bimonthly *Bulletin*. Seventy-seven new varieties of plants have evolved as a result of radiation-induced mutations, nearly 50 of which have been released to growers in the past 5 years. They include rice and wheat varieties with greater resistance to disease and to weather and with higher protein content and yield. One of the best-known flavors in the world—peppermint—might have been lost but for the use of the radiation method to develop resistance to a disease that threatened to destroy whole crops. Other methods altered the flavor to spearmint. The techniques have also affected the ornamental flower trade.

FOOD IRRADIATION IN CHILE

Chile is starting on a comprehensive study of radiation preservation of foods important in that country. Various government agencies—atomic energy, agriculture, fish, and public health—and the University of Chile are involved. Several international agencies are providing funds and technical assistance. Radiation facilities to be used include a 100-Ci ^{137}Cs source, a 0.8-MeV accelerator, a 22-in. cyclotron, and a 1-MW swimming-pool reactor as well as a ^{60}Co gamma source in Lima, Peru, for a joint fruit-fly irradiation study. Foods to be irradiated include potatoes for sprout inhibition; maize, wheat, and other grains for insect disinfestation; and fish and seafoods for length-

ening their storage life. Irradiation of blood and medical supplies will also be investigated.

RADIOISOTOPIC TIMEPIECE

Bulova Watch Co. is working on a completely new timepiece that is reported to use a small amount of alpha-emitting radioisotope as a time base. The nuclear watch, which this advanced research hopes to develop, would offer accuracy, achieved with a solid-state detector, and compactness, achieved by elimination of outside power sources. Since radioactive decay is not affected by temperature, the timepiece would not be affected by changes in ambient temperature. Such a system has a potential on-the-wrist accuracy of within a minute a year. It would be completely safe for a watchmaker or jeweler to handle, or for a person to wear, since the radioactive source would be enclosed in a metal shielding case.

At the present stage of the research, the working version, which displays time through an illuminated series of digits that report the time of day to 0.01 sec, is encased in a metal box about the size of a table-top television receiver. Bulova does not expect that new, sophisticated wristwatches will be available to consumers in the near future. The complex and extensive circuitry needed to count down from the high frequencies involved to seconds, minutes, and hours is still in the early development stages.

WOOD–PLASTICS MADE IN CANADA

Radiation Technology, Inc., has made its first shipment of irradiated wood–plastic flooring material to manufacturers of dimensional stock and parquet flooring. The material was produced in the St. Hilaire, Quebec, plant of Radiation Technology (Canada) Ltd.

PRICE OF WOOD–PLASTIC FLOORING ANNOUNCED

Atlantic Richfield has announced a price of $1.50 per square foot installed for its Permagrain, a wood–plastic composite made, using ^{60}Co irradiation, by its ARCO Chemical Division.

RADIATION PROCESSING OF WATER

The Ontario Water Resources Commission (135 St. Clair Ave. W., Toronto 7) is using two Gammacells (^{60}Co) supplied by Atomic Energy of Canada Limited to investigate the value of gamma irradiation of waste water to improve its settling ability and the biodegradability of some of its components. They are also studying gamma disinfection of both sewage and potable water. The radiation inactivation of several species of bacteria and viruses is being determined at different dose rates, and the effect of irradiation used in combination with other forms of water treatment will be compared with the effect of irradiation alone.

The Ontario Research Foundation (80 Bay St., Toronto) is using an AECL-supplied Gammacell to treat sewage. They have found that, if irradiated sewage is treated with lime to remove phosphates, the dissolved carbon content can be decreased about 50%. The effluent has less phosphorus but more dissolved carbon than that from conventional activated sludge plants. The Foundation has concluded that doses required to treat the sewage adequately are high and that great engineering problems would be encountered in a practical application of the technique.

IRRADIATED WAX–POLYETHYLENE BLENDS FOR APPLICATION TO SOLIDS

An improvement in wax–polyethylene blends which are particularly adapted for waterproofing of paper for cups, cartons, wrappers, and the like has been proposed in U. S. Patent 3,436,325. When paraffin or other hydrocarbon wax is blended with 2 to 25% polyethylene and then irradiated with doses of 2 to 200 Mrads from an electron accelerator, cross-linking is said to occur so as to produce a nearly water-white composition which has increased tensile strength, is uniform and compatible at both room and elevated temperatures, and may be applied without continuous heating or agitation. Greater surface area may be covered by the new product, and the coated surface has more seal strength, greater flexibility, and a higher gloss. A variety of uses is proposed: e.g., the impregnation of cloth, leather, and paper; the coating of metallic surfaces and textile fibers; and the insulation of electrical materials. (HW)

CYCLOTRON CORP. BUILDS 22-MeV MACHINE

The Cyclotron Corporation (950 Gilman St., Berkeley, Calif. 94710) has been awarded a contract to provide a 22-MeV cyclotron to the University of California at Los Angeles. The Model CS-22 compact accelerator will be installed at the UCLA Medical Center in August 1970 and will be used by the Laboratory of Nuclear Medicine and Radiation Biology

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in an extensive research and clinical program in nuclear medicine. The energetic beams of protons, deuterons, and helium ions from the machine bombard target materials to form unique radioactive isotopes, which are used to study body functions, diagnose abnormalities, and locate tumors. Funds for purchase of the cyclotron were provided by the Division of Biology and Medicine of the U. S. Atomic Energy Commission.

This will be the sixth cyclotron installation in a major U. S. hospital. The Corporation recently completed installation of a medical cyclotron at the Argonne Cancer Research Hospital in Chicago and is currently manufacturing an accelerator for the Mount Sinai Hospital in Miami Beach, Fla.

THE NUCLEAR INDUSTRY IN OHIO

The State of Ohio has published a report entitled "The Nuclear Industry in Ohio" which is intended to encourage and promote further utilization of nuclear energy by Ohio firms and to provide a broader understanding of the term "nuclear energy."

After starting out with an overview of the nuclear-chemical classification system and a nontechnical presentation of the principles of radioisotope applications, the report lists (1) Ohio manufacturers of nuclear-oriented equipment, (2) Ohio educational facilities that can be used for consulting, and (3) state and federal agencies involved in implementation of the nuclear program. Of particular interest is the description of the State atomic development activities, headed by Roger B. Williams, Coordinator, and under the surveillance of the Ohio Atomic Energy Advisory Board.

The report also includes a list of recommended federal publications and the depository libraries where they may be seen, a map showing the major federal nuclear facilities and operating contractors in Ohio, and a list of nuclear reactors and critical assemblies affecting Ohio.

The report is available for $1.00 from: Development Department, State of Ohio, 65 South Front Street, Columbus 43215. (PSB)

ARIZONA ATOMIC ENERGY COMMISSION NAMES GILBERT AS DIRECTOR

In November 1969 the AAEC [see *Isotopes and Radiation Technology*, 7(2): 246 (1969)] appointed Donald C. Gilbert as Director. Offices of the Commission are at 40 East Thomas Road, Suite 107, Phoenix, Ariz. 85012.

MIT TO OFFER SUMMER COURSE ON PHYSICAL ASPECTS OF NUCLEAR MEDICINE

Massachusetts Institute of Technology (MIT) has announced the scheduling of a 2-week summer course on *Physical Aspects of Nuclear Medicine*, July 20–31, 1970. The special program is an intensive course designed to bring participants up to date in the state of the art in nuclear medicine, with emphasis on physical aspects: new developments in isotope techniques, instrumentation, data processing, clinical applications, specific organ studies, and therapy. The course is sponsored jointly by MIT and Harvard Medical School. Some tuition grants will be available. For further information, contact Director of the Summer Session, MIT, Room E19-356, Cambridge, Mass. 02139.

FISSION-PRODUCT PALLADIUM

By F. P. Roberts*

A recent shipment to Oak Ridge National Laboratory of 10 g of fission-product palladium, for reshipment to The Netherlands for use in nuclear physics studies, culminated a successful effort by Battelle–Northwest scientists to isolate and purify this precious metal which exists in the waste from the Hanford nuclear fuel-processing plant.

Reactor-produced palladium is considered to be a potentially valuable by-product of the nuclear power industry.† It has important large-scale industrial uses as catalyst and in low-voltage switchgears in commercial equipment. Although the fission product is about 15 wt.% radioactive ^{107}Pd, commercial utilization may not be severely restricted because ^{107}Pd decays by emission of a very low-energy beta and no gamma, with a half-life of 7 million years. The specific activity is thus less than 100 μCi/g for the fission-product element. Availability of palladium from this source may reach 70 thousand oz/year by 1980, based on conservative projection of growth for the nuclear power industry.‡

The palladium recovered by Battelle–Northwest was isolated from stored alkaline Purex waste by

*Battelle Memorial Institute, Pacific Northwest Laboratory, Richland, Wash.

†C. A. Rohrmann, Large-Scale Exploitation of Fission-Product Rhodium and Palladium, *Isotop. Radiat. Technol.*, 6(4): 352-357 (Summer 1969).

‡C. A. Rohrmann, Value in Spent Fuel, *Isotop. Radiat. Technol.*, 6(1): 19-24 (Fall 1968).

absorption on a strong base anion exchanger. Both palladium and rhodium were present in these wastes as soluble anionic complexes of as yet undetermined composition and are strongly held by anion-exchange resin. The palladium was reclaimed by wet ashing of the resin and was purified and converted to palladium metal by well-known techniques.

The dose rate of the original 13 g of metal obtained at Battelle–Northwest was below the detection limits of conventional instruments. (REG)

INDUSTRIAL RADIOLOGY [RADIOGRAPHY]

By S. Rumyantsev; published in 1962, translated from the Russian by S. Semyonov and published in English in 1967 by Mir Publishers, Moscow (279 pp.).

Examples of industrial gamma radiography are discussed in detail—weld, ingot, and casting inspection, location of a defect and determining its size.

Following an explanation, as in all such Russian books, of the basic radiation principles involved, the types of photographic film used and developing times are discussed. Various data on gamma sources—^{60}Co ^{137}Cs, ^{192}Ir, ^{170}Tm, and others—are tabulated and the code designations of various Russian radiography units are identified. Methods of recording radiographs are also considered together with safety precautions that should be observed. (MG)

NEUTRON ACTIVATION DETERMINATION OF AGING CHANGES IN ONE-TIME CARBON PAPER*

Changes due to aging in one-time black carbon-paper ink were studied by neutron activation analysis. The ink, to which mercuric sulfide and cobalt naphthenate had been added for test purposes, was spread on ordinary carbon-paper backing, and the paper, with controls, was irradiated (5×10^{12} n/(cm^2)(sec) for 45 min. Samples of the irradiated paper were stored 3 days at 80 or 120°F under a pressure of 0.5, 2, or 4 psi to accelerate the aging process and were then used in typing. Mercury-203 and ^{60}Co in the carbon images were detected by gamma spectrometry in order to determine how much ink had been transferred from the carbon paper to the bond typing paper. A 3-day storage at 120°F simulates 1 year of storage under normal conditions, although this temperature could be reached in a freight shipment during the summer. (MG)

*Abstract of "Radiochemical Analysis of Composition Changes in One-Time Carbon Paper Due to Aging," TAPPI, 51: 508-511 (1968).

AMERICAN NUCLEAR SOCIETY 16th ANNUAL MEETING, LOS ANGELES, JUNE 29–JULY 2, 1970

A tentative program for the 16th ANS meeting in Los Angeles gives the dates on the Isotopes and Radiation sections as: Materials for Space Isotope Heat Sources, Capsule Materials and Heat Shield Materials on June 29; Methods of Analysis of Apollo 11 and other Extraterrestrial Materials, and Charged-Particle and Photon Analysis of Ultrapure Materials and Mineral Analysis, June 30; Large-Scale Radioisotope Production and Radiation Processing and Radiochemical Separations, July 1; Innovations in Instrumental Techniques and Methods, July 2. Also of interest to isotope people will be the session on Isotope Space Power Systems Technology on June 30 and the Ecology and Nuclear Education Session on July 1.

XUM

AEC Symposium Series

INHALATION CARCINOGENESIS

Proceedings of a Biology Division, Oak Ridge National Laboratory, conference, held at Gatlinburg, Tennessee, Oct. 8-11, 1969

Edited by M. G. Hanna, Jr., P. Nettesheim, and J. R. Gilbert

The deterioration of man's relationship with his environment is being documented by an increasing number of alarming findings. A positive correlation has been found to exist between the increased rate of lung cancer in humans and environmental air pollutants—such as motor vehicle exhaust, coal and asphalt dust, fumes from fuel oils, cigarettes, and other contaminants. The sponsoring agencies of this conference, the National Cancer Institute and the U. S. Atomic Energy Commission, felt that a conference on inhalation carcinogenesis would place the results of inhalation studies in proper perspective in the rapidly developing concern over environmental influences on man.

A basic need in the study of known and suspected carcinogens is a relevant model for testing, one which closely simulates the human mode of exposure. Inhalation systems are potentially able to fulfill these requirements. At this conference research was assessed to find out how well this need is being met.

A fundamental concern of the conference was evaluation of the importance and feasibility of using inhalation techniques to study respiratory carcinogenesis. Thus a considerable portion of the conference was devoted to the technology of inhalation studies. Chronic inhalation studies now under way were reviewed, and, finally, plans were presented for future programs in inhalation carcinogenesis.

Published as No. 18 in the AEC Symposium Series,
this book is available as CONF-691001 for $3.00 from

Clearinghouse for Federal Scientific and Technical Information
Springfield, Virginia 22151

RADIATION BIOLOGY OF THE FETAL AND JUVENILE MAMMAL

This volume contains the proceedings of the Ninth Annual Hanford Biology Symposium. The more than 75 papers included in this volume provide a comprehensive cross section of recent research on the metabolism of radionuclides and on the effects of radiation in the perinatal period. The areas covered include:

- Cross-placental transfer of radionuclides
- Mineral metabolism in the perinatal period
- Effects of radiation on the fetus and juvenile
- Response of the immature CNS to radiation
- Radiation effects in infants and children
- Mechanisms of differential sensitivity

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Index

Isotopes and Radiation Technology

Volume 7

A

Accelerators (Dynacote industrial electron)
availability of, 354
Accelerators (low-voltage)
use for production of neutrons for neutron radiography, 291–93
Activation analysis
(see Neutron activation analysis)
Advisory Committee on Isotopes and Radiation Development
membership appointment, 473
Aebersold, Paul C.
memorial lecture, 473
AECL Commercial Products
fabrication of ^{60}Co sources, 359
sales and development report, 360
Aerospace
radioisotope applications in, symposium on, 36–53
Aircraft
armor protection of, use of gamma radiography for studies of, 358
SST, inlet-shock-position sensing system for, use of avalanche semiconductor detectors in, 272, 274
tritium self-luminous exit signs for, loss of ^{3}H from, 349–52
American Chemical Society
educational awards for nuclear-oriented projects in chemistry and chemical technology, 362
Americium-241
availability, 123
use in calibrating radiation detectors, 438–39
use of radioisotopes in, survey of, 465–68
Apollo Lunar Surface Experiments Package
design of SNAP-27 system for, 443–47
Apollo Space Program
use of radioluminescent materials in, 64–67
Arizona
membership in the Western Interstate Nuclear Compact, 246
Arizona Atomic Energy Commission
director appointment of, 476
Australia
production and uses of radioisotopes in, 111–12
Aviation industry
use of radioisotopes in, Israeli booklet on, 463
Awards (Babcock–Hart)
recipient of, 246
Awards (undergraduate)
for nuclear-oriented projects in chemistry and chemical technology, 362

B

Barium-130
availability, 469
Barium-132
availability, 469
Barium-134
availability, 469
Barium-135
availability, 469
Barium-137
availability, 130, 469
Batteries (nuclear)
development by Donald W. Douglas Laboratories, 360–61
Beta gages
development for determination of weight loss in fabrics due to wear, 359
Beta irradiators
(see Irradiation facilities)
Beta particles
development of avalanche semiconductor detectors for, 270–73
Bibliographies
on production of radioisotopes in the IAN-R1 Reactor, 28
Biochemistry
Kyoto University gamma-irradiation facility for research in, 118–20
Biology
use of radioisotopes in, book on, 245
use of stable isotopes in, 371–75
Books and booklets
"Determination of Wear with Radioisotopes", 111
"Engineering Compendium on Radiation Shielding: Volume I, Shielding Fundamentals and Methods", 134
"Industrial Radiography", 477
"Isotopes in Chemistry", 118
"Nuclear Methods in the Aviation Industry", 463
"Radioisotopes and Inquiry", 245
Boron-10
availability, 469
Boron-11
availability, 469
Boron isotopes
operation of AEC's separation plant, contract extension for, 131
Brochures
"^{252}Cf: Its Use and Market Potential", 76
"Radioactive Isotopes in Industry", in German, 241

C

Cadmium-109
availability, 123
use in calibrating radiation detectors, 438
Cadmium-114
availability, 355
Californium-252
development of transportable source for neutron radiography, 132
loan agreements for, 471
newsletter on, availability of, 471
production program, 37
use as neutron source in neutron radiography, 62–63, 285–91
use as neutron source for in situ neutron activation of Ag, comparison with accelerators for, 411–18
use as neutron source for sulfur determination in coal, 131
use in oil-well logging, 245
uses and market potential of, Savannah River brochure on, 76
uses of, survey of, 306–16
Canada
gamma-irradiation synthesis in, 116–17
Carbon-13
availability of labeled compounds of, 123
price decrease, 354
Carbon-14
availability of labeled compounds of, 123
production in IAN-R1 Reactor, 28
Carbon paper
aging changes in, determination by neutron activation analysis, 477
Cardiac pacemaker
development of, 192–96
implantation in dog, 245
Cement
moisture content in, use of gamma radiation for measuring, 33
Ceramics
moisture content in, use of gamma radiation for measuring, 33

This index was prepared by Theodore F. Davis of AEC's Division of Technical Information Extension, Oak Ridge, Tenn.

XUM

Cerium-141
use in calibrating radiation detectors, 438
Cesium-137
availability, 123
price-increase proposal, cancellation of, 354
production in IAN-R1 Reactor, 28
use in calibrating radiation detectors, 438
use as gamma sources for determination of moisure content of various materials, 29–35
use in irradiation greenhouse at Kasetsart University, 114–15
Chemical technology
undergraduate awards for nuclear-oriented projects in, 362
Chemistry
applications of isotopes in, book on, 118
undergraduate awards for nuclear-oriented projects in, 362
use of stable isotopes in, 369–70, 375
Chile
food irradiation in, 474–75
Chlorine-37
availability, 130
Chromatography
(see Radiochromatography)
Chromium-50
availability, 469
Chromium-51
use in calibrating radiation detectors, 437
Chromium-54
availability, 469
Clams
radiopasteurization of, 78–79
Coal
analysis for ash by radiation-absorption technique, 464
analysis for sulfur by activation analysis, use of ^{252}Cf in, 131
Cobalt-60
availability, 123
availability of high-specific-activity for research on heat-source applications, 353–54
radiation source of, fabrication by AECL Commercial Products, 359
use for control of pipeline weld-inspection device, 76
use in calibrating radiation detectors, 438
use as gamma source for determination of moisture content of various materials, 29–35
use in Kyoto University medical irradiation facility, 118–20
use as power source in navigation light on St. Lawrence Seaway system, 359
use in radiometric viscometer, 75
use as radiotracer in measuring the abrasivity of magnetic recording tapes, 324
use in 250-thousand-curie irradiation facility at Lockheed-Georgia, 243–44
Concrete–polymer materials
applications of, 96–98
preparation of, 80–92
properties of, 92–96
Conferences and meetings
(see also Symposia)
on mutations in plant breeding, 107
on production and uses of radioisotopes in the Soviet Union, 124
on radiation preservation of food, 354
Crab meat
radiopasteurization of, 78–79
Curium-242
availability, 123
Cyclotrons
construction of 22-MeV at the University of California at Los Angeles, 475–76

D

Density
measurement of, use of avalanche semiconductors in, 272, 274
Donald W. Douglas Laboratories
development of nuclear battery, 360–61
Doors
intrusion of, use of radioisotopes for detection of, 339–40
Dynacote industrial electron accelerators
availability of, 354
Dysprosium-156
availability, 243
Dysprosium-158
availability, 469
Dysprosium-160
availability, 243
Dysprosium-162
availability, 243
Dysprosium-163
availability, 130
Dysprosium-164
availability, 469

E

Early Apollo Scientific Experiments Package
design of Apollo Lunar Radioisotopic Heaters for, 448–49
Earth sciences
use of stable isotopes in, 370–71, 375
East Germany
production of radioisotopes in, 462
Educational awards (undergraduate)
for nuclear-oriented projects in chemistry and chemical technology, 362
Electrolytic cells
mercury balance in, determination by isotopic-dilution technique, 463
Element-104
discovery of, 257–58
predicted properties of, 253, 255
Element-105
discovery of, preliminary information on, 258
predicted properties of, 253, 255
Element-106
predicted properties of, 253, 255
Element-107
predicted properties of, 253, 255
Element-108
predicted properties of, 253, 255
Element-109
predicted properties of, 253, 255
Element-110
predicted nuclear stability of, 259–60
predicted properties of, 253, 255
Element-111
predicted properties of, 253, 255
Element-112
predicted properties of, 253, 255
Element-113
predicted properties of, 253–55
Element-114
predicted nuclear stability of, 258–62
predicted properties of, 253–55
Element-115
predicted properties of, 254–55
Element-116
predicted properties of, 254–55
Element-117
predicted properties of, 254–55
Element-118
predicted properties of, 254–55
Element-119
predicted properties of, 254–55
Element-120
predicted properties of, 254–55
Element-121
predicted properties of, 255, 157
Element-122
predicted properties of, 257
Element-123
predicted properties of, 257
Element-126
predicted nuclear stability of, 259
Element-164
predicted nuclear stability of, 259
Elements (superheavy)
predicted properties of, 251–62
search for in nature, 262–63
synthesis of, methods for, 263–64
Engines (Stirling)
development for radioisotope-powered artificial hearts, 145–71
Engines (Rankine-cycle)
development for radioisotope-powered artificial hearts, 171–92
Equipment
marking with radioactive screws, 335–36
Erbium-167
availability, 469
Erbium-168
availability, 469
Erbium-170
availability, 469
Europium-152
availability, 123
Europium-154
availability, 123

F

Fabrics
weight loss due to wear of, development of beta gage for measurement of, 359
Films
"Atomic Revolution in Wood", 358
Fish
radiopasteurization of, 78–79
Fluid flow
measurement of, use of radiotracers in, 330–33
Food
(see also specific foods, e.g., Fish, Potatoes, etc.)
insect destruction in, by irradiation, 78
radiation preservation of, 77–79
radiation preservation of, conference on, 354
radiation preservation of, status of AEC and Army program for, 358
Food and Agriculture Organization
conference on radiation preservation of food, sponsorship of, 354
France
development of radioisotope power and heat generators, 99–107
production, sales, and use of radioisotopes in, 241
radioisotope catalog, 123
uses of radioisotopes in, survey of, 109–10
Fruit flies
control in the Mediterranean by radiosterilization, 474

G

Gadolinium-154
availability, 130
Gages (radiometric)
(see also Beta gages and Gamma gages)
aerospace applications of, 37–41
thickness measurement, 460
use in France, survey of, 110
Gallium-67
availability, 471
Gamma gages
use in determination of moisture in various materials, 29–35
Gamma radiation (low-energy)
use in industrial measurement and control devices, 381–94

XUM

Gamma radiography
design of portable ^{170}Tm unit for light metals, 458
Russian book on, 477
use in designing armor protection for aircraft, 358
uses in France, 110
Gamma sources
(see Irradiation facilities and specific Radioisotopes)
Georgia
regulatory authority over radioactive materials, 473
Germanium-71
use in calibrating radiation detectors, 438
Gold-198
production in IAN-R1 Reactor, 28
use in calibrating radiating detectors, 438
Goldblith, Samuel A.
recipient of Babcock–Hart Award, 246
Grain
destruction of insects in, by irradiation, 78
Turkish irradiator for, abandonment of, 132
Grenades (hand)
inspection of delay-charge element in fuses of, radiotracer technique for, 434–36

H

Hafnium-180
availability, 130
Hand grenades
inspection of delay-charge element in fuses of, radiotracer technique for, 434–36
Heart
development of radioisotope-powered pacemaker for, 192–96, 245
leakage diagnosis of, use of avalanche semiconductor detectors in, 274–76
Heart (radioisotope-powered)
development of a modified Stirling engine for, 161–71
development of a Rankine-cycle rotary expansion steam engine for, 182–92
development of a Rankine-cycle steam engine for, 171–81
development of a simplified Stirling engine for, 145–60
use of ^{147}Pm in, 141–44, 207–19, 231–40
use of ^{238}Pu in, 138–44, 197–206
Heat sources (radioisotopic)
(see Radioisotope-powered heaters)
Hittman Associates, Inc.
development of an isotopic heat source for use in heart-assist devices, 131
Hydrocarbons
radiation-chemical sulfonation of, economics of, 462

I

Idaho Nuclear Energy Commission
establishment and functions of, 132–34
India
production and use of radioisotopes in, 452–57
Indium-113m
availability of generators for, 123
Industry
use of radioisotopes in, German brochure on, 241
Information
storage of secret on foils, use of recoiled radioisotopes in, 335
Insects
destruction in foods by irradiation, 78
International Atomic Energy Agency
conference on radiation preservation of food, sponsorship of, 354
Iodine-123
new production method, 28
Iodine-125
availability, 123
use in calibrating radiation detectors, 438
use in miniature X-ray unit, 20–22
Iodine-131
production in IAN-R1 Reactor, 28
Iridium-192
availability, 123
Iron-54
availability, 469
Iron-55
use in calibrating radiation detectors, 437
use as X-ray source in diffraction analysis, 54–61
Iron-57
availability, 130
Iron-58
availability, 355
Irradiation facilities
availability of Gammacell 20, 125
availability of Scholastic Radatron, 470–71
^{137}Cs greenhouse at the Kasetsart University, 114–15
availability of ^{90}Sr–^{90}Y, 353
construction of dual-purpose research and production complex by Radiation Technology, Inc., 246
demonstration of Hydroponica at Nuclex 69, 459–60
demonstration of Koltos at Nuclex 69, 460
description of 250-thousand-curie ^{60}Co at Lockheed-Georgia, 243–44
description of Kyoto University ^{60}Co for biomedical research, 118–20
design of gamma using spent fuel elements, 461
development by Radiation Technology, Inc., 134–35, 246
Turkish grain, abandonment of, 132
development of transportable beta for medical uses, 107
Irradiation sources
(see Irradiation facilities and specific Radioisotopes)
Isotope Products Laboratories of Los Angeles
availability of sealed sources of radioisotopes from, 247
Isotope targets
production at ORNL, 1–19
Isotopes
(see Radioisotopes and Stable isotopes)
Isotopes Information Center
publications of, 247–49
"Isotopes and Radiation Technology"
summary of replies to reader interest check sheet, 125–30

J

Japan
Kyoto University gamma-irradiation facility for biochemical research, 118–20
production and use of radioisotopes, 121–23

K

Kasetsart University
^{137}Cs gamma-irradiation greenhouse at, 114–15
Krypton-85
availability, 123
enrichment of, conceptual design of a continuous thermal-diffusion plant for, 23
use as tracer for detection of cracks in turbine blades, 277–84
use in radioisotope-powered self-illuminating trafic signs, 472
Kyoto University
gamma-irradiation facility for biochemical research, 118–20

L

Labeled compounds
analysis by chromatography, 70–72
availability from CEA-CEN de Saclay, 123
biosynthesis of, 69–70
peptide synthesis, 70
self-radiolysis of, 72
tritium biochemicals, preparation by modified Wilzbach technique, 241–42
tritium-labeling techniques, 68–69
Labels
use of radioisotopes for solid surfaces, 334–35
Lanthanum-138
availability, 130, 243
Law enforcement
use of radioisotopes in, 334–40
Lead-204
availability, 130
Lead-206
availability, 243
Lead-207
availability, 130
Lead-208
availability, 469
Licensing and regulations
on uses of radioisotopes, changes in, 356–57
Lighthouses
design of radioisotope powered in Sweden, 108–9
Lignin
radiation-induced polymerization with aldehydes and vinyl monomers, Canadian development of, 117
Lobsters
radiopasteurization of, 78–79
Lockheed-Georgia
description of 250-thousand-curie ^{60}Co irradiation facility, 243–44
Locks
use of gamma-ray spectrometry and radioactive key in, 337, 339
Lubricants
radiation-resistant, Canadian development of, 117
Luminescent materials
(see Radioluminescent materials)

M

Magnesium-25
availability, 243
Magnesium-26
availability, 243
Magnetic recording tapes
abrasivity of, radiotracer method for measuring, 324
Magnetosphere
measurement of, use of avalanche semiconductor detectors in, 272
Mangoes
destruction of insects in by irradiation, 78
Marine products
radiopasteurization of, 78–79
Marking
of equipment, use of radioactive screws in, 335–36

XUM

of hydrogenous surfaces with ^{3}H, 336
of solid surfaces, use of radioisotopes in, 334–35
Mars
life-detection system for, use of avalanche semiconductor detectors in, 272
Massachusetts Institute of Technology
course on physical aspects of nuclear medicine, 476
Materials
determination of moisture in, by gamma radiation, 29–35
labeling of surfaces of, use of radioisotopes in, 334–35
McMaster University, Hamilton, Ont.
nuclear engineering program, 470
Meats
(see also specific meats, e.g., Fish, Lobsters, Poultry, etc.)
moisture content in, use of gamma radiation for measuring, 33
radiopasteurization, 78–79
radiosterilization of, 79
Medicine (nuclear)
development of transportable beta irradiator for use in, 107
developments in radioisotopic diagnoses with no administration of radioactive materials, 473–74
physical aspects of, MIT course on, 476
recognition as a special field for physicians, 473
use of avalanche semiconductor detectors in, 274–76
use of stable isotopes in, 371–75
Mercury
determination in electrolytic cells by isotopic-dilution technique, 463
Mercury-196
availability, 130, 469
Mercury-203
use in calibrating radiation detectors, 438
Minerals
exploration for, use of ^{252}Cf in, 411–18
Moisture
(see Water)
Moleculon
expansion in radiation developments, 135
Molybdenum-98
availability, 355, 469
Mutations
in plant breeding, conference on, 107
in plants, production by irradiation, 474

N

National Bureau of Standards
issuance of radioactive standards, 354–55
Navigational light
use of ^{60}Co as power source for, 359–60
Neodymium-146
availability, 130
Neutron activation analysis
use in studying aging changes in one-time carbon paper, 477
use in France, survey of, 110
use in identification of soil specimens, 319–24
use of ^{252}Cf as neutron sources for, 131, 245, 315–16, 411–18
Neutron gages
use in the determination of moisture in roof sandwiches, 325–30
use and performance in the determination of moisture in soils, report on survey of, 297–305
Neutron radiography
development of transportable source of ^{252}Cf for, 132
facility at the University of Texas TRIGA Reactor, 294–98
use of ^{252}Cf in, 62–63, 132, 285–91
use of low-voltage accelerator neutron source in, 291
uses in France, 110
Neutron sources
availability of ^{124}Sb–Be from AECL commercial products, 470
use of ^{252}Cf for, 62–63, 132, 245, 285–91, 306–16, 411–18
use of low-voltage accelerator for, 291–93
Nickel-62
availability, 469
Nondestructive testing
aerospace applications of, 47–48
North Dakota
regulatory authority over radioactive materials, 355
Nuclear batteries
development by Donald W. Douglas Laboratories, 360–61
Nuclear engineering
education program at McMaster University, 470
Nuclear Materials and Equipment Corporation
operation of AEC's boron-isotope separation plant, contract extension, 131
Nuclear medicine
(see Medicine (nuclear))

O

Oak Ridge Associated Universities
training activities of the Special Training Division of, review, 402–7
Oak Ridge National Laboratory
availability of stable isotopes from, 130, 243, 355, 469
sale of 1 million curies of ^{90}Sr, 131
Oak Ridge National Laboratory Isotope Target Center
description of facilities and capabilities of, 1–19
Ohio
nuclear industry in, report on, 476
Oil (heavy)
heat value of, determination by neutron attenuation, 464
"Old Bremsstrahlung University"
training activities in the nuclear sciences, review of, 402–7
Onions
sprouting of, inhibition by irradiation, 77–78
Oysters
radiopasteurization of, 78–79

P

Pakistan
production of radioisotopes in, 341–42
Palladium
recovery from Hanford wastes, 476–77
Palladium-102
availability, 130
Palladium-104
availability, 130
Palladium-105
availability, 130
Palladium-106
availability, 130
Palladium-110
availability, 130
Papayas
destruction of insects in, by irradiation, 78
Peat
moisture content in, use of gamma radiation for measuring, 33
Periodic Table
prospects for further considerable extension of, 251–64
Pho/Gamma Camera
use in nuclear medicine, 358
Phosphorus-32
availability, 123
availability of labeled compounds of, 123
production in IAN-R1 Reactor, 28
Physics (nonnuclear)
use of stable isotopes in, 368–69, 375
Physics (nuclear)
use of stable isotopes in, 365–68, 374–75
Pipelines
weld-inspection device for, use of ^{60}Co for control of, 76
Plants
mutations in breeding of, conference on, 107
nutrition studies of, use of radiochromatogram scanner and tracer experiments in, 343–48
Plastic–wood combination
(see Wood–plastic combination)
Canadian development of, 116–17
development of Skinpreg Process for, 242
film on, 358
use in AEC Germantown Headquarters, 358
Plutonium
detection in wounds, use of avalanche semiconductor detectors for, 275–76
Plutonium-238
standard source of, availability of, 355
study as power source for artificial heart, 138–44, 197–206
Plutonium-242
availability of high-purity, 353
Polonium-210
availability, 123
price increase, 469
Polyethylene foam
production of cross-linked by irradiation, new company established for, 362
Polyorganosiloxanes
radiation-induced synthesis of, Canadian development of, 117
Potatoes
sprouting of, inhibition by irradiation, 77–78
Poultry
radiopasteurization of, 78–79
Promethium-147
study as power source for artificial heart, 141–44, 207–19, 231–40
use in radioluminescent devices for space vehicles, 64–67
Promethium oxalate ($Pm_2(C_2O_4)_3.3H_2O$)
calcining to Pm_2O_3, thermal analysis of, 24–27
Pumps
use of radiotracers to measure discharge rates of, 332–33

R

Radiation detectors
aerospace applications of, 36–37, 42–45
calibration of, use of radioisotopes in, 436–39
Radiation detectors (semiconductor, avalanche)
development for low-energy radioactivity, 266–77
Radiation Dynamics, Inc.
sale of Dynacote industrial electron accelerators, 354
Radiation Machinery Corporation
developments at, report on, 359
Radiation processing
activities at Radiation Technology, Inc., 358–59
activities in Canada, 116–17

economics of sulfonation of hydrocarbons, 462
of wax–polyethylene for waterproofing of paper cups, 475
Radiation shielding
engineering compendium on, 134
Radiation sources
(see Irradiation facilities and specific radioisotopes)
Radiation Technology, Inc.
construction of research and production irradiation facility, 246
development of new irradiation facilities, 134–35
radiation-processing activities, 358–59
Radioactive materials
regulatory authority by states, 355, 473
Radioactivity
low-energy, development of avalanche semiconductor detectors for, 266–77
Radioactivity standards
availability of ^{238}Pu, 355
availability of ^{226}Ra, 355
availability of ^{228}Th–^{208}Tl, 355
availability of ^{113}Sn–^{113}In, 355
survey on status and needs for, 135
Radiochromatography
use in plant nutrition studies, 343–48
Radiography
(see Gamma radiography and Neutron radiography)
Radioisotope power generators
Beta-C, design of, 459
development for underwater applications, 449–51
Efer, design of, 459
French development of, 99–107
Gisete 6, design of, 459
Ripple VI, design of, 108
SNAP-3, performance of, 356
SNAP-19, use in Nimbus B2 weather satellite, 131
SNAP-21, development for ocean exploration devices, 356
SNAP-19, purchase by AEC, 472
SNAP-27, design for Apollo Lunar Surface Experiments Package, 443–47
Radioisotope-powered heaters
aerospace applications, 45–47
availability of high-specific-activity ^{60}Co for research on, 353–54
design for the Early Apollo Scientific Experiments Package, 448–49
development for artificial heart engines, 131, 137–241
development for moon landing experiments, 132
French development of, 104–6
Radioisotopes
(see also specific radioisotopes)
aerospace applications of, symposium on, 36–53
availability of sealed sources of, 247
British catalog of, 123
French catalog of, 123
licensing and regulations on uses of, changes in, 356–57
potential uses for security and law enforcement, 334–40
production in East Germany, 462
production in the IAN-R1 Reactor, 28
production in Pakistan, 341–42
production, sales, and use of, in France, 241
production and uses in Australia, 111–12
production and uses in India, 452–57
production and uses in Japan, brochure on, 121–23
production and uses in South Africa, 113
production and uses in the Soviet Union, conference on, 124
sales for 1968, 360
use in aviation, Israeli booklet on, 463
use in chemistry, book on, 118
use in determining wear in machine parts, book on, 111
use in France, survey of, 109–10
use in Latin America, survey of, 465–68
use in security and law enforcement, 334–40
uses of, consultant services for, 361, 469
Radioluminescent materials
loss of ^{3}H from aircraft exit signs, 349–52
use in Apollo space program, 64–67
use in France, survey of, 110
use in traffic signs, 472
Radiopasteurization
of foods, 77–79
of foods, conference on, 354
of foods in Chile, 474–75
of foods, status of AEC and Army program for, 358
Radiopharmaceuticals
manufacturer for, establishment of new, 359
Radiosterilization
of fruit flies in the Mediterranean, 474
of human tissues for transplant surgery, 247
of meats, 79
Radiotracer techniques
(see Tracer techniques)
Radium-226
standard source of, availability of, 355
Rankine-cycle engines
development for radioisotope-powered artificial hearts, 171–92
Reactor fuel elements
use of spent as gamma irradiator, 461
Reactors (IAN-R1)
production of radioisotopes in, 28
Reactors (PARR)
radioisotope production in, 341–42
Reactors (University of Texas TRIGA)
neutron radiography facility at, 294–96
Roof sandwiches
water determination in, use of neutron moisture gage for, 325–30
Russia
production and uses of radioisotopes in, conference on, 124
Ruthenium-96
availability, 130
Ruthenium-102
availability, 130

S

Safe locks
use of gamma-ray spectrometry and radioactive key in, 337, 339
Samarium-148
availability, 130
Samarium-154
availability, 130
Sanders Nuclear Corporation of Nashua
investigation of ^{170}Tm oxide as fuel for isotopic-power devices, contract extension for, 355–56
Scallops
radiopasteurization of, 78–79
Sea foods
radiopasteurization of, 78–79
Security
use of radioisotopes in, 334–40
Selenium-74
availability, 243
Selenium-76
availability, 243
Selenium-77
availability, 469
Self-luminescent aircraft exit signs
loss of ^{3}H from, 349–52
Semiconductor detectors (avalanche)
development for low-energy radioactivity, 266–77
Sewage water
radiation processing of, 475
Shielding
(see Radiation shielding)
Shock waves
measurement of, use of avalanche semiconductor detectors in, 272, 274–75
Shrimp
radiopasteurization of, 78–79
Silicate (foamy)
moisture content in, use of gamma radiation for measuring, 33
Silicon-28
availability, 130
Silver
in situ neutron activation of, comparison of ^{252}Cf and accelerators as neutron sources for, 411–18
Sodium-24
production in IAN-R1 Reactor, 28
Soils
identification of, use of neutron activation analysis in, 319–24
moisture content in, use of gamma radiation for measuring, 29–32
moisture determination in, report on survey of neutron gages for, 297–305
South Africa
production and uses of radioisotopes in, 113
South Carolina
regulatory authority over radioactive materials, 355
Soviet Union
production and uses of radioisotopes in, conference on, 124
Space vehicles
use of radioluminescent materials in, 64–67
Stable isotopes
availability from ORNL, 130, 243, 355, 469
national uses and needs for, Ad Hoc Panel report on, 363–77
Stainless steel (316)
corrosion in H_2SO_4, radioactive tracer technique for studying, 407–10
States
regulatory authority over radioactive materials, 355, 469
Steel
production of, use of radioisotopes in, 419–33
Steel (mild)
corrosion in H_2SO_4, radioactive tracer technique for studying, 407–10
Stirling engines
development for radioisotope-powered artificial hearts, 145–71
Strawberries
radiopasteurization of, 78
Strontium-90
sale of 1-million-curie order by ORNL, 131
Sulfur
determination in coal, use of ^{252}Cf in, 131
Sulfur-33
availability, 469
Sulfur-34
availability, 469
Sulfur-35
availability of labeled compounds of, 123
production in IAN-R1 Reactor, 28
Sweden
design of radioisotope-powered lighthouse, 108–9
Symposia
(see also Conferences and meetings)
on aerospace applications of radioisotopes, 36–53
Systems for Nuclear Auxiliary Power
(see Radioisotope-powered generators)

XUM

T

Tapes (magnetic recording)
  abrasivity of, radiotracer method for measuring, 324
Technetium-99*m*
  availability of generators for, 123
Tellurium-120
  availability, 469
Tellurium-122
  availability, 130
Tellurium-123
  availability, 130
Tellurium-124
  availability, 243
Texas, University of
  TRIGA Reactor, neutron radiography facility at, 294–96
Textile fibers
  moisture content in, use of gamma radiation for measuring, 29–32
Textiles
  irradiation grafting of, Canadian development of, 116
Thallium-205
  availability, 130
Thailand
  description of the ^{137}Cs gamma-irradiation greenhouse at the Kasetsart University, 114–15
Thorium-228
  standard source of, availability of, 355
Thrusters
  leak-detection systems for, use of avalanche semiconductor detectors in, 272
Thulium-170
  investigation as fuel for isotopic-power devices, Sanders Nuclear Corporation contract extension for, 355–56
  use in opposed-arc-weld-inspection system, 440–42
  use in portable gamma-radiography unit for light metals, 458
Thulium-171
  study as power source for artificial heart, 142–44, 222–31
Time measurement
  development of nuclear device for, 475
Tin-113
  standard source of, availability of, 355
Tin-114
  availability, 130
Tin-117
  availability, 130
Tissues
  radiosterilization for transplant surgery, 247
Titanium-47
  availability, 130
Tracer techniques
  trilium detection in, use of avalanche semiconductor detectors for, 274
  use in detection of cracks in turbine blades, 277–84
  use in France, survey of, 110
  use in measuring abrasivity of magnetic recording tapes, 324
  use in measuring corrosion rates of mild steel and 316 stainless steel in H_2SO_4, 407–10
  use in measuring discharge rates of high-head turbines and pumps, 330–33
  use in measuring length and weight of delay-charge elements in hand-grenade fuses, 434–36
  use in measuring wear in machine parts, book on, 111
  use in measuring water movement in trees, 396–401
  use in melt-shop steelmaking operations, 419–33
Traffic signs
  testing of radioluminescent, 472
Training courses
  "Introduction to Radioisotopes" at the Post-Graduate Education Centre, AERE, Harwell, England, 470
Transactinide elements
  predicted properties of, 251–62
  search for in nature, 262–63
  synthesis of, methods for, 263–64
Trees
  grain determination using radioisotopes, 245
  water movement in, radioisotope procedures for measuring, 396–401
Tritium
  availability of labeled compounds of, 123
  loss from luminescent aircraft exit signs, 349–52
  preparation of labeled compounds of, 68–69, 241–42
  use in tracer-techniques development of avalanche semiconductor detectors for, 274
Tungsten-180
  recovery and purification of electromagnetically separated, 378–80
Tungsten-182
  recovery and purification of electromagnetically separated, 378–80
Tungsten-183
  recovery and purification of electromagnetically separated, 378–80
Tungsten-184
  recovery and purification of electromagnetically separated, 378–80
Tungsten-186
  recovery and purification of electromagnetically separated, 378–80
Turbine blades
  cracks in leading edge of, use of ^{85}Kr in detection of, 277–84
Turbines
  use of radiotracers to measure discharge rates of, 330–31
Turkey
  development of grain irradiator, cancellation of, 132

U

United Kingdom
  radioisotope catalog, 123
U. S. Army Cincinnati Procurement Agency
  development of beta gage for measuring weight loss of fabrics due to wear, 359
Uranium-233
  availability of high-purity, 353
Uranium-234
  availability of high-purity, 353

V

Vegetables
  dehydrated, improvement of rehydration time by irradiation, 79
Viscometers
  design of radiometric, 75
Voltek, Inc.
  production of cross-linked polyethylene foam, 361–62

W

Water
  determination in roof sandwiches, use of neutron gage for, 325–30
  determination in soils by neutron gage, report on survey of, 297–305
  determination in various materials by gamma radiation, 29–35
  movement in trees, radioisotope procedures for measuring, 396–401
Water (waste)
  radiation processing, 475
Wax–polyethylene blends
  radiation processing, 475
Wear
  determination in floor covering using radioisotopes, 125
  determination in machine parts using radioisotopes, book on, 111
  determination in magnetic recording tapes, 324
Welds (opposed-arc)
  gamma-ray inspection system for, 440–42
Western Interstate Nuclear Compact membership, 246
Wheat
  destruction of insects in by irradiation, 78
Wood
  moisture content in, use of gamma radiation for measuring, 33
Wood–plastic combination
  Canadian development of, 116–17
  commercial contract for, 132
  development of Skinpreg process for, 242
  film on, 358
  production by Radiation Machinery Corp., expansion of facilities for, 359
  price of, 475
  sale of, 475
  use in AEC Germantown Headquarters, 358
Wounds
  locating of Pu in, development of avalanche semiconductor detectors for, 275–76

X

X radiation (low-energy)
  use in industrial measurement and control devices, 381–94
X-ray-diffraction analysis
  use of beta-excited and electron-capture X-ray sources in, 54–61
X-ray fluorescence analysis
  use of avalanche semiconductor detectors in, 269–71
  use in determining wear in floor covering, 125
X-ray sources
  production from radioactive isotopes for diffraction analysis, 54–61
X-ray unit
  use of ^{125}I in miniature, 20–22

Y

Ytterbium-169
  availability, 123
Yttrium-90
  availability, 123

Z

Zinc-64
  availability, 130
Zinc-67
  availability, 355

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